# Desalination and Water Treatment www.deswater.com

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# Novel pilot plant-scale graywater treatment system using titanium ball, membrane and advanced oxidation process

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Received 1 October 2008; Accepted in revised form 2 September 2009

# ABSTRACT

In this study, graywater, which is defined as all household wastewater originating from a source other than the toilet, was treated through a process combining titanium ball, microfiltration membrane and advanced oxidation treatment. Removal efficiency of COD, suspended solids, turbidity, color and *E. coli* was studied to determine the possibility of reuse of the graywater as water for fire fighting, irrigation, toilet and car washing. The pH was 7–7.7. The removal efficiency of each factor using titanium ball was as follows: COD 90%, suspended solids 92%, turbidity 98%, color 95%, and *E. coli* 2%. Removal efficiency of each factor using a microfiltration membrane was as follows: COD 98%, suspended solids 96%, turbidity 100%, color 98%, and *E. coli* 30%. As a subsequent process, removal efficiency of each factor by using an advanced oxidation process was as follows: COD 99%, suspended solids 100%, turbidity 100%, color 100%, and *E. coli* 100%. The quality of the treated graywater was sufficient to establish a sustainable water circulation system to reuse apartment wastewater. The TB (titanium ball)–membrane–OP system satisfied the standards for the reuse of water in Korea, which are as follows: COD of less than 20 mg/L, turbidity of less than 2 NTU, color of less than 20 Pt-Co, suspended solids of less than 5 mg/L, no detection of *E. coli*.

Keywords: Graywater; Titanium ball; Membrane; Advanced oxidation process

# 1. Introduction

The interest in wastewater recycling has recently increased due to a growth in water demand, shortages of water due to low rainfall, and various other economic and environmental factors [1]. The reuse of graywater is being spotlighted as a means of water conservation. Graywater is generally defined as all household wastewater excluding toilet effluent. However, it is a frequent misconception that graywater is cleaner than combined domestic wastewater, and therefore can be reused with minimal or no treatment [2]. In fact, graywater can contain various pollutants such as suspended solids, organic matter, nutrients and detergents, with turbidity of 15–240 NTU, COD (chemical oxygen demand) of 180–650 mg/L, 5–15 mg/L TKN (total Kjeldahl nitrogen) and TP (total phosphorus), and 1–30 mg/L anionic detergents [3–8]. A wide variety of treatment systems have been reported in the literature, and these vary greatly in terms of their complexity and the degree of treatment provided [9–17]. Ramon et al. [18] studied a method of graywater treatment

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Presented at EuroMed 2008, Desalination for Clean Water and Energy Cooperation among Mediterranean Countries of Europe and the MENA Region, 9–13 November 2008, King Hussein Bin Talal Convention Center, Dead Sea, Jordan.

using an ultrafiltration membrane and a nanofiltration membrane. Garland et al. [19] studied a method of graywater treatment using hydroponic plants, while Gross et al. [2] studied a method of graywater treatment using a vertical flow constructed wetland. Even when reused for purposes other than drinking and bathing, such as for fire fighting, irrigation, and water for the toilet and car washing, a certain quality standard for reused water must be satisfied. In Korea, the quality standard for reused water is as follows: COD of less than 20 mg/L, turbidity of less than 2 NTU, color of less than 20 Pt-Co, suspended solids of less than 5 mg/L, and zero *E. coli*.

The objectives of this study are to evaluate the titanium ball tower-MF (microfiltration) membrane–OP (oxidation process) system for the removal of COD, turbidity, color, suspended solids, and *E. coli* in graywater treatment.

# 2. Materials and method

# 2.1. Sampling

The area of each apartment in the complex studied was 55.45 m<sup>2</sup>, and 170 apartments were studied in total. The apartment complex studied is located in Bundang-Gu, Kyungki-Do, Korea.

#### 2.2. Analysis methods

pH was determined using a glass electrode pH meter (Orion, Model 525A). COD, turbidity and color were measured using the HACH digestion vials (HACH, DR/2012). Measurement of MLSS (mixed liquid suspended solids) was done according to the 2540D method in Standard Methods for the Examination of Water and Wastewater, 20th edition, Clescerl et al., (1998). *E. coli* was estimated using CHROMagar TM ECC. To detect *E. coli*, the following method was used: First, the specimen was inoculated directly on the surface of the medium. Second, the plate was incubated at 35±2°C for 18–24 h.

## 2.3. Titanium ball-MF membrane-OP system

# 2.3.1. Titanium ball-MF membrane system

Fig. 1 represents the experimental system used in this study, which consists of a titanium ball tower and an aerobic submerged MF membrane. The volume of the sediment reactor was 1 m<sup>3</sup>, and the volume of the titanium ball reactor was 3 m<sup>3</sup>. The microorganisms in the titanium ball were capable of removing organic matter. The volume of the MF membrane reactor was 1 m<sup>3</sup>, and the volume of the storage tank was 1 m<sup>3</sup>. During 90 days of the experiment, total HRT (hybrid retention time) was maintained at 12 h (6 h/sediment reactor, 4 h/ titanium ball reactor, 2 h/MBR). The module type of the membrane was hollow fiber, and its pore size was  $0.4 \,\mu$ m. The material was polyvinyl difluoride. The operation type of membrane was the submerged type, and urethane and epoxy resin were used for bonding. The model name of the membrane was SuperMAK®.

### 2.3.2. Disinfection system

Fig. 2 shows the ozone experimental system used in this study. This process was performed after the graywater had passed through the titanium ball-MF membrane system. The ozone reactor consisted of an ozone generator, an ozone detector, a stainless steel reactor, and equipment to destroy the remaining ozone. Ozone contacted the graywater in the cylindrical batch reactor by bubbling. The reactor was made of stainless steel to avoid oxidation. A ceramic air distributor was located at the bottom of the cylinder, and introduced ozone into the reactor. Ozone was generated by flowing oxygen into the ozone generator. The ozone concentrations at different O<sub>2</sub> flow rates and output voltages are listed in Table 1. The results of the experiments were estimated at 10 min, 20 min, and 30 min after contacting ozone. To determine optimal  $O_2$ flow rates and output voltages in terms of the efficiency of the removal of color, turbidity and organic matter, a



Fig. 1. Schematic diagram of titanium ball-MF membrane system.

Table 1 Characteristics of ozone concentration according to the  $O_2$  flow rate and output voltage

O <sub>2</sub> flow rate (Nm <sup>3</sup> /m)	Output voltage (kV)	O <sub>3</sub> amount (g/h)
1.5	6	2.6
	7	4.0
2	6	2.6
	7	4.8
2.5	6	2.4
	7	3.7
	8	5.4
3	7	3.2
	8	5.2



Fig. 2. Schematic diagram of the ozone system.

total of nine experiments were performed, and 2.5  $\text{Nm}^3/\text{m}$  and 7 kV were determined to be the optimal O<sub>2</sub> flow rate and output voltage, respectively, as these showed good efficiency compared to other operating conditions.

# 3. Results and discussion

# 3.1. Evaluation of titanium ball –MF membrane–oxidation process system

Fig. 3a shows the efficiency of COD removal by the titanium ball–MF membrane–oxidation process system. The maximum COD level in the influent was 187 mg/L (63rd day), and the minimum value of influent was 80 mg/L. Removal efficiency of COD was about 84–100%.

Although the removal efficiency was 84% on the 28th day, the actual level of COD in effluent was 14 mg/L. As the legal standard for water reuse is less than 20 mg COD/L in Korea, this value satisfies the legal standard. By the 42nd day, the efficiency of COD removal was 100%. Ramon et al. [18] have conducted experiments on low strength graywater treatment using an ultrafiltration (UF) membrane and a nanofiltration (NF) membrane. Depending on the membrane used, UF removed 45-70% of COD, as well as 92–97% of turbidity. Depending on the membrane used, NF removed 93.3% of COD, as well as 98.1% of turbidity. Figs. 3b and 3c show the efficiency of the removal of turbidity and suspended solids by the titanium ball-MF membrane-oxidation process system. Maximum value of influent was 220 NTU, and the minimum value of influent was 34 NTU. The maximum value of suspended solids in the influent was 181 mg/L, while the minimum value was 30 mg/L. Through treatment combining the titanium ball and the MF membrane, suspended solids were removed very effectively. The efficiency of turbidity removal was 98–100%. Fouling of the MF membrane did not occur in this study. Fouling is a major issue in the membrane process. For example, Lodge [10] compared wastewater with graywater in UF following biological treatment and found similar fouling, reasoning that the difference could be explained due to the higher SS concentration in the wastewater in that study. In this study, as suspended solids were removed effectively through the titanium ball process, it seems that hardly any fouling of the MF membrane occurred. Fig. 3d shows the efficiency of color removal by the titanium ball-MF membrane-oxidation process system. The maximum color value of influent was 520 Pt-Co, and the minimum value of influent was 165 Pt-Co. Through treatment with the sediment reactor and MF membrane, organic matter, suspended sludge and DOM (dissolved organic matter) were removed effectively. The efficiency of color removal was greater than 90% from the 5th day. The decolorization of wastewater using various membrane-based, pressure-driven processes has been investigated in several studies [20–23]. MF and UF are low-pressure membrane processes that separate suspended solids, macromolecules and colloids from a feed stream [10,22]. As a pretreatment step, MF can significantly enhance water quality for a reasonable cost. Decolorizing textile wastewater [15], municipal wastewater and groundwater supplies [3] are some examples of the application of membranes in this area.

The efficiency of E. coli removal was 100%. Although the titanium ball–MF membrane system could not efficiently remove *E. coli* (13 CFU/mL), the oxidation process system showed good treatment in terms of removing *E. coli* (0 CFU/mL). In addition, *E. coli* was removed within 0–15 min in the oxidation process system.



(c) Efficiency of suspended solids removal

(d) Efficiency of color removal

Fig. 3. Efficiency of COD, turbidity, suspended solids and color removal.

# 4. Conclusion

The results reported in this study indicate that the titanium ball–MF membrane–OP system shows good treatment capacity. The titanium ball–MF membrane system studied was able to effectively remove COD, turbidity, color, and suspended solids, while the OP was effective for the removal of *E. coli*. As the MF membrane could not completely remove *E. coli*, a disinfection system was required. If graywater is reused for a human-contact purpose, a disinfection system has to be installed. The titanium ball–MF membrane–OP system satisfies Korean

standards [26] for the reuse of water, which are: COD less than 20 mg/L, turbidity less than 2 NTU, color concentration less than 20 Pt-Co, suspended solids less than 5 mg/L, and zero detection of *E. coli*. Since the MF membrane was very effective for the treatment of COD, turbidity, color and suspended solids, it was unnecessary to employ UF membranes or NF membranes. The MF membrane was also more economical than the UF membrane or the NF membrane. Friedler et al. [14] reported that if the practice of on-site graywater reuse becomes widespread, the cost of such systems will inevitably decrease, making them more appealing to individual consumers. In addition,

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under typical conditions on-site graywater reuse is a feasible solution for decreasing overall urban water demand, not only from an environmental standpoint, but also in economic terms. Treated graywater can be used as water for the toilet, for fire fighting, for car washing, for irrigation, and for constructed wetlands and ponds. Prathaper et al. [25] reported that it would be beneficial to individuals and to society if the building industry can be persuaded to install graywater treatment systems in (a) new houses, (b) new apartment complexes, and (c) public buildings, such as mosques and schools, where existing plumbing could easily be modified to separate graywater from blackwater. Through such measures, a sustainable water circulation system can ultimately be accomplished.

## Acknowledgement

This work was supported by Yonsei University, Center for Future Infrastructure System, a Brain Korea 21 program, Korea.

This research was supported by a grant (06 Construction Core B02) from Construction Core Technology Program funded by Ministry of Construction and Transportation of the Korean government.

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