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Co-treatment systems combined with unit processes for dye wastewater recycling

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A B S T R A C T

In this paper, co-treatment systems were configured to recycle dye wastewater by combining unit processes such as membrane separation, ozone oxidation and electrolysis. In our previous study of cotreatment with membrane separation and ozone processes (modes 1–3), it was found that ozone oxidation with enriched oxygen was effective in removing dye wastewater. Additional two co-treatment systems (mode 4: membrane separation and electrolysis and mode 5: mode 4 + ozone process with enriched oxygen) were configured in this paper and operated continuously for 24 h. Compared to our previous results, co-treatment systems with electrolysis were found to further enhance the removal efficiencies of organic matter, T-N and T-P by 4, 20 and 15%, respectively. Especially, their color removal efficiencies were about 75–85 and 93% with modes 4 and 5, respectively, indicating that co-treatment with electrolysis is an excellent system in color removal.

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1. Introduction

Within the proliferation and increasing complexity of industrial production facilities, the amount of process wastewater that is discharged has substantially increased, polluting water systems and making river water difficult to use as a water resource without significant purification. This leads to elevated water treatment cost and the deployment of advanced treatment technologies is required. Reflecting this situation, recycling technologies for process wastewater from production processes have been widely developed as approaches to mitigate water shortage and securing water resources. Since process wastewater from a given production process is generally quite stable with respect to pollutant compositions and their concentrations, there is little fluctuation in mass loading rates, consequently allowing consistent, steady and predictable treatment if an appropriate process is employed. The benefits of recycling are well established, particularly where suppliers and consumers of process water are located in close proximity [\[1–3\].](#page--1-0)

After amendments to the water supply and waterworks installation act and administrative regulations in South Korea, a water reclamation and reuse system is actively encouraged to be

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installed in the production facilities nationally. Accordingly, many domestic industries in South Korea are constructing or already operating plants to reuse process wastewater. However, such treatment systems are limited to their own internal purposes, with a knowledge and experience base not publically available, and not developed for general purposes. Therefore, to extend the open access availability of water reuse technologies, it is necessary to obtain individual technology elements for pre-treatment and advanced treatment, and to remodel them for general practical use through systematic research and development. In this regard, cotreatment systems combining existing technology elements provide a number of alternative options that may be applied for different wastewater processing requirements.

The process wastewater from the textile/dye industry, in particular, contains non-degradable organic matter with high alkalinity, chromaticity and concentration, complicating its treatment processes for reuse $[4-6]$. Combinations of chemical treatment (coagulation and fenton oxidation) and biological treatment (standard activated sludge method) have generally been applied to treat dye wastewater, but proven to be inadequate in terms of color removal, as well as giving rise to a large volume of sludge [\[7–9\].](#page--1-0)

As a pre-treatment step a biological treatment process with fluidizing media, was used on the dye wastewater as an initial stage. This serves as a replacement for the activated sludge processes which are currently used. The treated water was then

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further processed in co-treatment systems. The conventional biological treatments yield sludge bulking and provide for insufficient in color removal for azo-type dyes [\[12\]](#page--1-0), Consequently, alternative and more effective biological treatment processes such as using fluidizing media are necessary. This is known to reduce the amount of sludge production and pollutant concentrations significantly in the discharged water [\[10\]](#page--1-0) while providing comparable other outcomes to conventional activated sludge processes. This low sludge rate better matches enforced environmental regulations in South Korea, such as the regulation of total process waste emissions. It is worth noting that consistent criteria regarding color removal have not been established [\[11\]](#page--1-0).

In this paper, the stages following the biological pretreatment (with fluidizing media) are examined, to bring the dye wastewater quality to a level suitable for reuse. We combined a separation membrane process (which is very effective in removal of organic matter and suspended solids) $[13,14]$, an ozone process (which has strong oxidation potential and is excellent in disinfection, deodorization and decoloration) [\[15\]](#page--1-0) and an electrolysis process (which is effective in decoloration and color removal), and operated in continuous fashion to demonstrate the system for provision of treated recycled water.

The experiments follow up our previous study [\[16\]](#page--1-0) and compare 5 different configurations:

Mode 1: membrane only

Mode 2: membrane followed by ozone generator fed with air Mode 3: membrane followed by ozone generator fed with enriched air $(32\% \text{ O}_2)$

Mode 4: membrane followed by electrolysis

Mode 5: membrane followed by electrolysis followed by ozone generator fed with enriched air $(32\% O₂)$

Modes 1–3 were reported previously [\[16\]](#page--1-0), but have been further tested and assessed here to provide direct comparison against an additional advanced treatment system for dye wastewater treatment, namely the enhanced effect of the electrolysis process on color removal.

2. Experimental

Table 1

2.1. Simulated dye wastewater

Discharged water from a biological treatment process with fluidizing media was analyzed over a period of 6 months in terms of COD_{Mn} , COD_{Cr} , T-N, T-P, Cl⁻ and SS. The maximum concentrations were then used to make synthetic wastewater (170.70, 404.29, 23.27, 1.06, 258.26 and 240.0 ppm, respectively) so that cotreatment systems would be tested with the worst case of water quality. For simulated dye wastewater, 10 ppm of each disperse dye (black, scarlet and violet), which are commonly used in dye industry [\[9\]](#page--1-0), was added to the synthetic wastewater, as listed in Table 1 [\[16\]](#page--1-0).

2.2. Process design and configuration of co-treatment systems

2.2.1. Configuration of modes 1, 2 and 3

Co-treatment systems were configured with a separation membrane process (mode 1) and an additional ozone process (modes 2 and 3). They were operated according to the procedures outlined in our previous study $[16]$. The separation membrane was made of PVDF (polyvinylidene fluoride) hollow fiber modules (6 m²/module). It was operated intermittently at 7 min/run and 3 min/stop, providing over a 24 h operation period a treatment capacity of 1 m^3 /day. Backwashing was conducted once a week with NaOCl solution (100–500 mg/L) for 30 min. There was 90 min of stabilization time before another continuous operation started. The ozone process had two options (mode 2: air injection and mode 3: enriched oxygen injection). An ozone generator was designed for a capacity of 10 g/h and a pure oxygen generator (Fineteckwin, South Korea) was used to blend 32% oxygen enriched air.

2.2.2. Configuration of modes 4 and 5

Mode 4 consists of separation membrane and electrolysis processes. For mode 5, an ozone process with enriched oxygen injection was added to mode 4. The schematic diagram of the electrolysis reactor and the configuration of electrodes are shown in Figs. 1 and 2, respectively. [Fig.](#page--1-0) 3 depicts the overall co-treatment systems with the electrolysis process.

The electrolysis reactor used a direct current power supply with a capacity of 30 V and 30 A. To avoid electrode corrosion caused by chlorine gas $(Cl₂)$ generation and electron emission from the electrode, we used mesh type insoluble anode that had iridium oxide (IrO₂) electrodeposited on titanium (Ti). The cathode, where reduction reactions take place, was made of mesh type stainless steel (SUS). An important consideration is the separation between the anode and cathode. When the distance between anode and cathode is short, the electric current increases, leading to an increase in the removal efficiency while the current efficiency decreases, giving rise to more power consumption. If the electrode distance is long, organic matter removal and inorganic oxidation can be reduced, requiring high voltage. Thus, reported optimum conditions [\[17,3\]](#page--1-0), which are the distance of 6 mm and the current density of 1.0 $A/dm^2/L$ over an immersed area of electrode, were adopted in this paper. Also, a direct oxidation zone (18 L) and an indirect one (45 L), which are left and right zones in the electrolysis reactor, respectively, were installed to maximize the efficiency of electrolytic treatment. The purpose was to extend the residence

Electrolysis Reactor

Fig. 1. Schematic diagram of electrolysis reactor.

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