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Coupled electron beam radiation and MBR treatment of textile wastewater containing polyvinyl alcohol

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HIGHLIGHTS

- We use electron beam coupled with an MBR to treat textile effluent containing PVA.
- Radiation products of the organic wastewater were well utilized by microbes in MBR.
- COD was steadily removed by the electron beam radiation-MBR treatment.

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ABSTRACT

Advanced oxidation processes (AOP) can be combined with biological treatments for recalcitrant organic pollutant decomposition. However, there has been no thorough investigation on the coupling of AOPs and membrane bioreactors (MBR) to treat polymer organic pollutants. This study proposes a new AOP that couples electron beam (EB) radiation and MBR treatment. This method was applied to treat real textile effluents containing polyvinyl alcohol (PVA). During the stable operation stage, 31 ± 7% (n = 28) COD was removed by the EB-MBR process. COD removal was enhanced to 45% at the end of the research period without process optimization. In addition, both the membrane flux and activated sludge system exhibited good stability. Only a 2% membrane flux decreased was observed after a 46 d operation period. PVA radiolysis and biofacies analysis mechanisms are also discussed. By contrast, PVA degradation using only the MBR treatment was ineffective in this study. This ineffectiveness was caused by membrane interception and floccule formation by PVA and activated sludge.

1. Introduction

Dyes and sizes are two main types of recalcitrant organic pollutants in textile wastewater. Dyes, especially azo dyes, make up approximately 70% of all dyestuffs by weight (Singh and Arora, 2011). Their intermediates, e.g., aromatic amines, are toxic, carcinogenic and mutagenic, posing a potential health hazard to human beings (Dumont et al., 2010; Turesky, 2005). Polyvinyl alcohol (PVA) is an excellent sizing agent that is widely used by various industries. The global annual PVA consumption has surpassed 1,000,000 tons (Yoshio and Henry, 2010). However, the BOD_5/COD ratio of PVA (1.5 g/L) is only 0.064 (Lin and Lo, 1997). The biodegradability is dependent on microbe adaptation, but not degradation (Chen et al., 2000). For textile wastewaters containing PVA and dyes, traditional biological treatments are usually ineffective. Recently, combining advanced oxidation process (AOP) and biological treatments has become a feasible strategy for recalcitrant organic pollutant decomposition and refractory organic wastewater treatment (Sun et al., 2013b). Ionizing radiation, i.e., electron beam radiation and gamma radiation, is a special AOP. It can both produce hydroxyl radicals (•OH) and generate reducing species, such as hydrogen atoms (H•) and hydrated electrons (e^-_{aq}) (Wang and Wang, 2007). Previously, biodegradability enhancement was achieved in textile wastewater using electron beam radiation combined with an

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activated sludge process (Mohd Nasir et al., 2010). The biodegradability ratio of non-radiated wastewater was 0.34–0.61, which increased to 0.87–0.96 after electron beam radiation. Additionally, biodegradability was improved in desizing wastewater containing PVA using gamma radiation (Jo et al., 2006). In addition, gamma radiation could enhance the biodegradability of PVA in aqueous solution (Sun et al., 2012) and azo dye in aqueous solution (Sun et al., 2013a). Because of its excellent performance for textile wastewater treatment, a 10,000 m^3/d dyeing wastewater treatment plant created and effectively applied an electron beam-biological process (Han et al., 2012). However, little research has been performed regarding the treatment of PVA-polluted textile wastewater using the coupled process. This paper implements the application of a combined electron beam radiation-membrane bioreactor treatment (EB-MBR). This technique is applied to PVA-polluted textile wastewaters in this work.

2. Materials and methods

2.1. EB-MBR process

The EB-MBR process consisted of batch EB pretreatment and continuous MBR treatment. The radiated wastewater from the electron beam was continuously pumped to an MBR using a peristaltic pump. As a controlled trial, the raw, non-radiated water was directly treated using another MBR. All conditions of the two MBRs were the same. The EB radiation used a 10 MeV electron accelerator (Rhodotron TT 200, IBA, Belgium). The accelerator was controlled by an automatic control system that could alter the current intensity (0–10 mA), scan length (0–100 cm) and flow velocity of the wastewater (0–12 m/min). The absorbed dose was quantified using a film dosimeter (B3 WINdose, GEX, USA). In general, there was no need to enter the radiation chamber. In addition, the accelerator could cut off the power supply at any time. After power off, it did not emit radiation.

The maximum flux of the submerged MBRs (height 60 cm, diameter 11 cm and working volume 4.3 L) with hollow fiber membrane (PVDF, 0.02 μm) was 12.5 L/(m^2 h). Activated sludge (MLSS 1.29 g/L, MLVSS 0.93 g/L) sampled from an aeration tank in a textile wastewater treatment plant (Suzhou, China) was used in the MBRs. The hydraulic retention time (HRT) of 24 h was controlled by a peristaltic pump (BT100-2J, Longer, China) by adjusting the effluent.

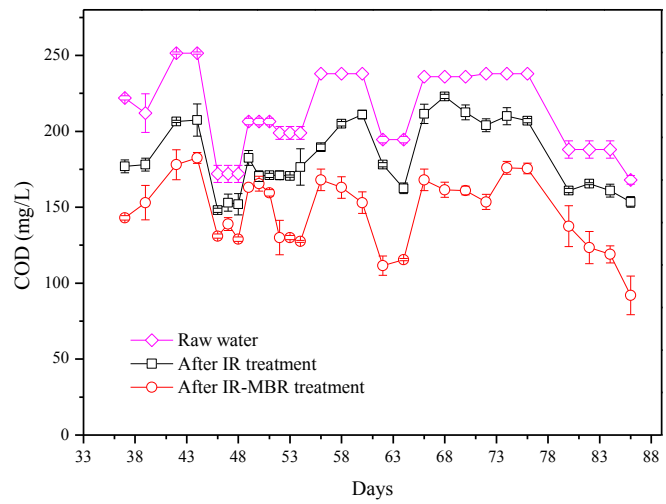

Fig. 2. Effect of the electron beam radiation-MBR coupling process on the COD removal efficiency (Temp., DO, pH and ORP of activated sludge were 19.9 ± 1.1 °C, 2.15 ± 0.08 mg/L, 8.39 ± 0.14, 147 ± 36 mV, respectively, for n = 11).

2.2. Wastewater and chemicals

The raw water was composed of textile wastewater (plant effluent from Suzhou, China) and added PVA (the average polymerization degree was 1700 ± 200, CP) with 211.7 ± 26.3 mg/L (n = 28) of COD, 90–100 mg/L of PVA, and pH of 8.1 ± 0.2 (n = 7). In addition, a 0.1‰ nutrient solution was added. The nutrient solution contained 2.80 g/L KH_2PO_4, 1.32 g/L $MgSO_4$, 150 g/L NH_4Cl, 7.2 g/L $NaNO_3$, 0.03 g/L $FeCl_3$, 0.8 g/L $CaCl_2$, 0.02 g/L $CuCl_2 \cdot 4H_2O$, 0.04 g/L $MnCl_2 \cdot 4H_2O$, 0.26 g/L $CoCl_2 \cdot 6H_2O$, 0.2 mg/L folic acid and 0.69 mg/L nicotinic acid.

Iodine (AP), potassium iodide (AP), boric acid (AP) and other chemicals were all purchased from commercial channels. All reagents and chemicals were used without further purification. All samples were prepared in deionized water.

2.3. Analytical methods

PVA concentration was analyzed by using a visible spectrophotometer (722s, Shanghai Precision & Scientific Instrument Co.,

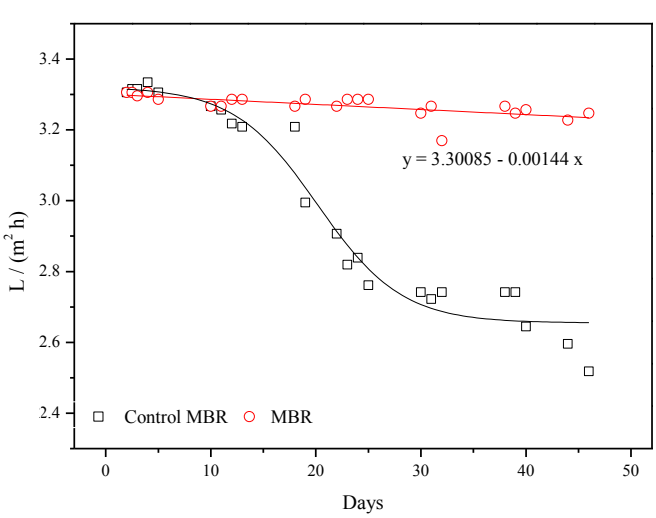

Fig. 1. Membrane flux trends of the MBR and controlled MBR.

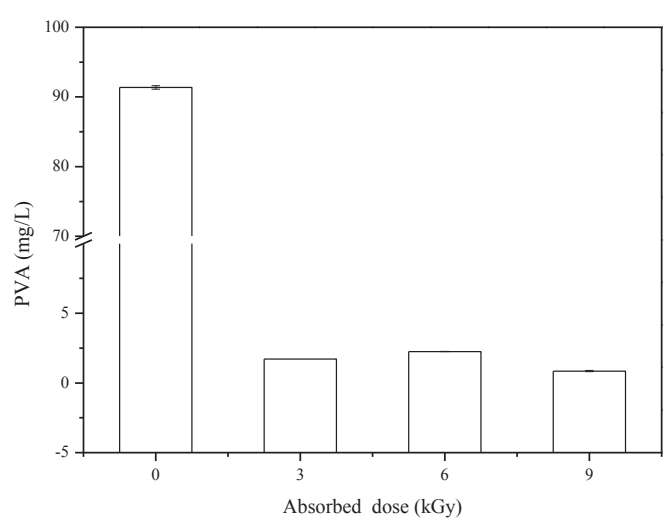

Fig. 3. Variation of PVA degradation based on the absorbed dose.