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Remediation of wastewater from pulp and paper mill industry by the electrochemical technique

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ABSTRACT

This research attempted to find the optimum condition for color and total COD reduction in wastewater from the pulp and paper mill industry by using electrocoagulation techniques in batch and continuous modes. Six pieces of iron plates constructed in parallel configurations were used as electrodes. The effect of key parameters including the type of polyelectrolyte, current density, initial pH of the wastewater, and the circulating flow rate of wastewater in the reactor were investigated. The results indicated that the polyelectrolyte had no effect on pollutant removal. At optimum conditions, greater than 97% of color and 77% of total COD were effectively removed with a total operating cost of approximately 0.29 USD/m^3 wastewater. First order rate kinetics best explained the reduction of color and total COD concentration, the model fitting the actual data very well. For the continuous mode, the treatment process reached the steady state condition within 2.15 h and the efficiency of color and total COD reduction was greater than 91% and 77%, respectively. The properties of wastewater including color, total COD, BOD_5, TSS, TDS, pH and iron ions content were in the range of the acceptable values of current Thai Government standards.

1. Introduction

The pulp and paper industry is an almost global, being present in most developed and developing countries and is one of the most important industries in Thailand because it has the potential to compete with other countries due to the plentiful supply of sustainable renewable fast growing forest resources and water supplies. However, this plant utilizes a high quantity of water, between 76–227 m^3 per ton of product, resulting in large amounts of wastewater generation and concomitant economic and ecological/environmental costs/problems. Indeed, such wastewater contains a large amount of pollutants characterized by biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids (SS), toxicity, and colorants which cause bacterial and algal slime growths, thermal impacts, scum formation, color problems, and a loss of both biodiversity and aesthetic beauty in the environment [1]. The main treatment processes used at pulp and paper mill plants are primary clarification (sedimentation or flotation), secondary treatment (activated sludge process or anaerobic digestion) and/or tertiary processes (membrane processes as ultrafiltration)[2]. Activated sludge plants have been the most common wastewater treatment process for the removal of organics in our country; however, there are several problems with the process. It produces sludges with very variable settlement properties, it is sensitive to shock loading and toxicity, and its capacity to remove poorly biodegradable toxic substances is limited. Therefore, many researchers attempted to develop the new technologies for complementing or even replace some of these treatments. The white rot fungus, *Ceriporiopsis subvermispora* CZ-3, was shown to be effective for reducing the concentration of pollutants in the effluent from a pulp bleaching process, with approximately 90% color, 45% total COD, 62% lignin and 32% adsorbable organically bound halogens (AOX) removal by the fungus within 48 h at an operating temperature and pH of 30–35°C and 4.0–4.5, respectively, in the presence of 1 g/L glucose [3]. However, in the economically and environmentally more desirable absence of glucose, the fungus could only remove color up to 62%. Subsequently, Amat et al. [4] used solar energy together with different types of catalyst, such as Fenton agent and TiO_2, to degrade pollutants from board paper industries and revealed that the Fenton reagent gave a higher degree of total COD and BOD_5 depletion compared to TiO_2. In the same year, Wong et al. [5] reported on the flocculation performances of nine cationic and anionic polyacrylamides, each with different molecular weights and charge densities, for treating pulp and paper mill wastewater. They revealed that cationic polyacrylamides, like Organopol 5415, with a very high molecular weight and low charge density achieved a reduction of 95, 98 and 93 % in turbidity, total suspended solid (TSS) and total COD, respectively, with a sludge

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Nomenclature

$(\text{total COD})_t$	total chemical oxygen demand at time t (g O_2/L)
$(\text{total COD})_{t+\Delta t}$	total chemical oxygen demand at time $t+\Delta t$ (g O_2/L)
F	Faraday's constant (26.8 Ah)
I	current intensity (A)
L	volume of electrolyte (L)
V	cell voltage (V)
t	electrolysis time (h)

volume index (SVI) of 14 mL/g at the optimum dosage of 5 mg/L. In a different approach, Buzzini et al. [6] reported a system based upon precipitation with a microbial community which provided approximately 80–86 % and 75–78 % total COD removal with and without recirculation of the effluent, respectively. By using the combined processes of coagulation-flocculation and heterogeneous photocatalysis to treat the post-bleaching effluent from a cellulose and paper industry, Rodrigues et al. [7] reported that the optimal conditions for coagulation were pH 6.0, 80 mg/L of $FeCl_3 \cdot 6H_2O$, and 50 mg/L of chitosan while the optimized photocatalysis conditions were found at pH 3.0 in 0.50 g/L of TiO_2 and 10 mmol/l of H_2O_2. Effluent turbidity decreased sharply after coagulations (from 10 FTU of in natura samples to 2.5 FTU without chitosan and 1.1 FTU with chitosan) and a decrease in the concentrations of N-ammoniac, N-organic, nitrate, nitrite, phosphate, and sulfate ions after coagulation. This combined method (coagulation followed by photocatalysis) resulted in a biodegradability index of 0.71, transparency, and absence of color and odor in the treated water.

However, without disregarding the already summarized treatment techniques, one of the most interesting and effective processes for treating some pollutants in wastewater are the electrochemical process because of their convenience and typically greater effectiveness than the above traditional methods [8]. Electrochemical processes can be applied in various configurations such as electrooxidation, electroprecipitation, electrocoagulation, electrodeposition, electro-Fenton, etc. Using three-dimensional electrodes with a $Ti/Co/SnO_2–Sb_2O_5$ anode to treat paper mill wastewater, Wang et al. [9] reported that pH and current density had a significant effect on the total COD and color removal efficiency. Removal of chromium and organic pollutants simultaneously by electroprecipitation lead to good (approximately 98%) chromium removal within 60 min but whilst the organic pollutants contained in wastewater such as oil and grease, color and the level of BOD_5, total COD and total kjeldahl nitrogen (TKN) were also markedly reduced [10], they were still higher than the acceptable values of the government standard. However, using electrocoagulation instead of electroprecipitation was found to be effective at both reducing the amount of chromium (within 10 min) together with up to 96 % of the organic pollutants present in the wastewater [11].

In the studies reported here, importantly, real wastewater from a pulp and paper mill plant, instead of using synthetic wastewater as in many previous works, was treated by the electrocoagulation technique both in batch and continuous processes. The effects of various parameters on color and total COD removal were explored to optimize the efficiency and cost effectiveness of the system.

2. Experimental

The experiment was carried out in a bench-scale laboratory at ambient temperature both in batch (Fig. 1(a)) and continuous modes (Fig. 1(b)) by employing actual wastewater from

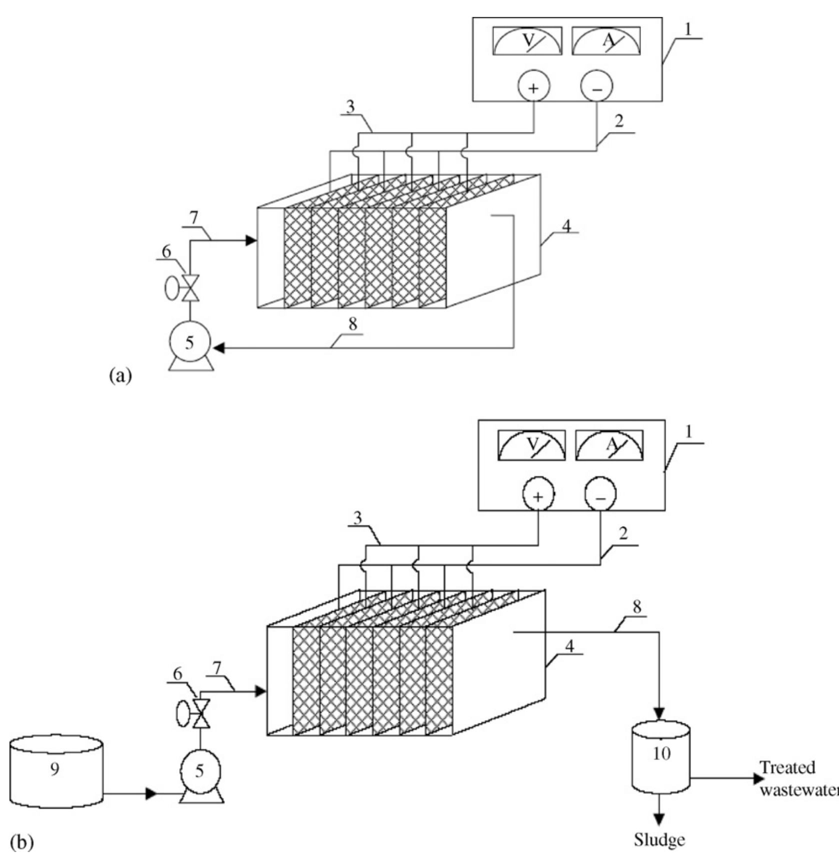

Fig. 1. Experimental set up for batch (a) and continuous modes (b): (1) power supply; (2) cathode; (3) anode; (4) reactor; (5) pump; (6) valve; (7) inlet stream; (8) outlet stream; (9) wastewater reservoir tank; (10) filtration tank.