Journal of the Taiwan Institute of Chemical Engineers 42 (2011) 380–386

Contents lists available at ScienceDirect

Journal of the Taiwan Institute of Chemical Engineers

journal homepage: www.elsevier.com/locate/jtice

Biotreatment of a triphenylmethane dye solution using a Xanthophyta alga: Modeling of key factors by neural network

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ARTICLE INFO

Article history:
Received 12 May 2010
Received in revised form 20 July 2010
Accepted 6 August 2010
Available online 26 November 2010

Keywords:
Biodegradation
ANN modeling
Textile dye
Macroalgae
Decolorization

ABSTRACT

In this paper biotreatment of triphenylmethane dye, Malachite Green (MG), by a Xanthophyta alga, *Vaucheria* species, was investigated. The results obtained from batch experiments revealed the ability of *Vaucheria* sp. to remove MG. The effects of operational parameters such as initial dye concentration, temperature, pH and algal amount on biological decolorization efficiency were examined. The results showed that the biological decolorization efficiency decreased with increasing initial MG concentration. The decolorization rate also enhanced with increasing the temperature, initial pH of the dye solution and the amount of biomass rose. Biological treatment of MG solution by live and dead alga was compared. The reusability and efficiency of the live alga in long-term repetitive operations were also examined. The batch experiments results revealed the ability of algal species in biological degradation of the dye. An artificial neural network (ANN) model was developed to predict the biological decolorization of MG solution. The findings indicated that artificial neural network provided reasonable predictive performance ($R^2 = 0.979$). The influence of each parameter on the variable studied was assessed, and reaction time and initial dye concentration were found to be the most significant factors, followed by initial pH, amount of alga and temperature.

1. Introduction

The removal of hazardous compounds from industrial effluents is a growing need at the present time (Mittal *et al.*, 2005). Dyes are synthetic, aromatic, water soluble, dispersible, organic colorants, having potential application in various industries. The large quantity of aqueous waste generated by textile industries has become a significant environmental problem (Khataee *et al.*, 2009a). Furthermore, effluent from the dyeing industry is one of the most problematic wastewaters to be treated, not only because of its high chemical and biological oxygen demand, suspended solids and content of toxic, carcinogenic, mutagenic or teratogenic compounds, but also its color, which is visually the most noticeable contaminant (Aksu, 2005; Chang *et al.*, 2000; Khataee *et al.*, 2009a). So, the removal of dyes from aqueous effluent has received considerable attention within environmental research (Khataee, 2009b). Ozonation, photooxidation, electrocoagulation, adsorption, activated carbon, froth flotation, reverse osmosis, ion exchange, membrane filtration and flocculation processes, are applied for color removal from textile effluents. Some of these techniques have been shown to be effective, although they have some limitations (Daneshvar *et al.*, 2004; Khataee *et al.*, 2009c; Mittal *et al.*, 2005; Robinson *et al.*, 2001).

In recent years, a number of studies have focused on some micro/macro-organisms which are able to biodegrade or bioaccumulate the dyes in wastewaters (Adav *et al.*, 2009; Aksu, 2005; Aksu and Tezer, 2005; Chang and Kuo, 2000; Chang *et al.*, 2000; Daneshvar *et al.*, 2007a; You and Teng, 2009). Biological decolorization of triphenylmethane dye such as Malachite Green has focused primarily on the decolorization of dyes by reduction reactions to its respective derivatives. Decolorization of organic dyes occurs under anaerobic (methanogenic), anoxic and aerobic conditions by different trophic groups of bacteria (Pandey *et al.*, 2007).

Algae are photosynthetic organisms, which are distributed in nearly all parts of the world and in all kinds of habitats. Algae can degrade a number of dyes, postulating that the reduction appears to be related to the molecular structure of the dyes and the species of algae used. Algae species, being highly versatile, have developed enzyme systems for the decolorization and mineralization of organic dyes under certain environmental conditions. Many algae displayed their effectiveness in degradation and decolorizing of azo dyes in wastewater effluents (Ertugrul *et al.*, 2008; Wang *et al.*, 2007). Acuner and Dilek (2004) and Jinqi and Houtian (1992) stated that some species of *Chlorella pyrenoidosa* and *Chlorella*

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doi:10.1016/j.jtice.2010.08.006

vulgaris degraded azo dyes and decolorized dye wastewater. They also found that some algae could utilize aniline, a degradation product of azo dye breakdown. Mohan *et al.* (2002) also investigated the green algae belonging to *Spirogyra* species as viable biomaterials for biological treatment of simulated synthetic azo dyes effluents. Lei *et al.* (2002) showed the ability of seven microalgae belonging to green and blue-green algae to remove pyrene from solution either by bioaccumulation or biotransformation, which were species-dependent. El-Sheekh *et al.* (2009) reported the ability of seven algae (*i.e. C. vulgaris*, *Lyngbya lagerlerimi*, *Nostoc lincki*, *Oscillatoria rubescens*, *Elkatothrix viridis* and *Volvox aureus*) to decolorize and remove methyl red, orange II, G-Red (FN-3G), basic cationic, and basic fuchsin dyes.

In this work, macroalga *Vaucheria* sp. was used in order to decolorize a dye solution containing Malachite Green. Malachite Green is a triphenylmethane dye which most commonly used for the dyeing of cotton, silk, paper, leather and also in manufacturing of paints and printing inks (Gupta *et al.*, 2004). Biological removal of MG in the presence of different microorganisms has been reported previously (Bekci *et al.*, 2009; Daneshvar *et al.*, 2007a,b; Jadhav and Govindwar, 2006; Khataee *et al.*, 2009d, 2010; Kumar *et al.*, 2006; Parshetti *et al.*, 2006). However, to the best of our knowledge, the application of macroalga *Vaucheria* sp. for biological treatment of MG has not been reported.

Therefore, this study aims to investigate the potential of macroalga *Vaucheria* sp. for decolorization of the solution containing MG. The effects of operational parameters such as initial dye concentration, initial pH, temperature and amount of algal on color removal efficiency have been also investigated. In addition, the reusability of the alga during repetitive decolorization operations was examined.

Another aspect of this work was the development of a multilayer feed forward neural network model to predict the biological decolorization efficiency of MG. The major challenge in modeling of bioprocesses is the nonlinear and time-varying nature of such processes (Rani and Rao, 1999). The ANN based models can estimate any nonlinear function for an unknown system while in design of experiments (DOE) (*e.g.* response surface methodology) the form of function must be selected (polynomial) by user and it needs some knowledge about the system. Thus, the application of an artificial neural network was tried to predict the performance of the biological process. One of the characteristics of modeling based on artificial neural networks is that it does not require the mathematical description of the phenomena involved in the process, and might be useful in simulating complex biological systems (Despange and Massart, 1998; Khataee and Mirzajani, 2010).

2. Materials and methods

2.1. Algal biomass

The algal species was acquired from Azna-lake in North of Iran. The algal species was washed with distilled water to remove macro/microscopic contaminations. According to its morphology and microscopic observation, it was identified as *Vaucheria* species belongs to Xanthophyta (Fig. 1(a)). The thallus consists of a branched, aseptate, coenocytic filament with apical growth. The numerous chloroplasts are oval or elliptical without pyrenoids. The branching is irregular. Cross-walls are not present to separate the cells of the filament and the branches. Thus the protoplasm is continuous along its entire length and extends without a break into the branches. The filament wall is relatively thin and weak. It consists of an inner layer of cellulose and an outer of pectic substances (Vashishta *et al.*, 2005). The microscopic picture was taken Olympus BX41 microscope (Japan).

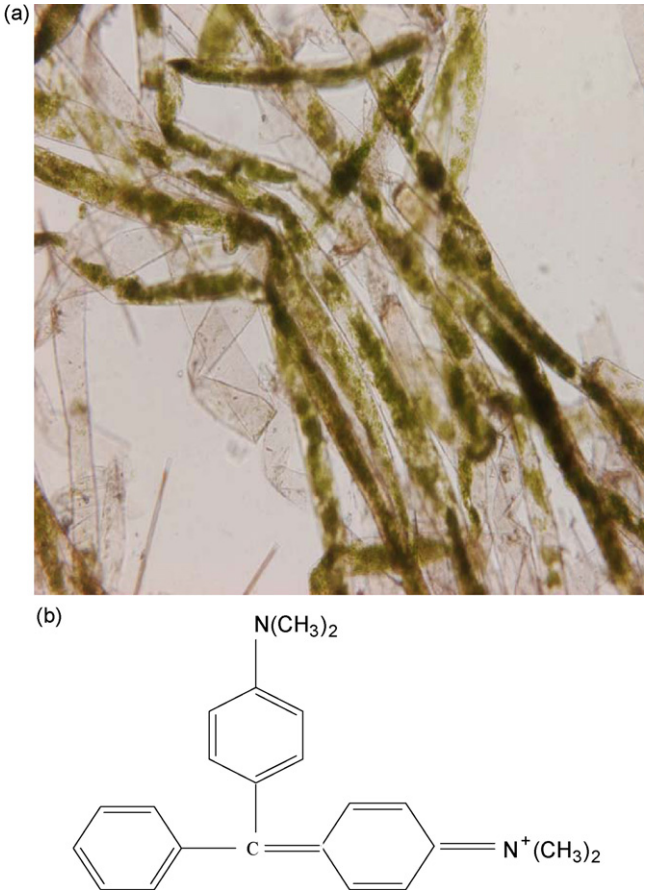

Fig. 1. (a) Light microscopic picture of *Vaucheria* sp.; (b) chemical structure of Malachite Green.

2.2. Instrumental analysis

The triphenylmethane dye, Malachite Green (Fig. 1(b)), was purchased from Merck (C.I. 42000, λ_{max} 919 nm). At regular time intervals of biotreatment process, samples were taken and the remaining MG was determined with a spectrophotometer (UV/Vis spectrophotometer WPA Lightwave S2000, England) at maximum absorption wavelengths, λ_{max} = 619 nm and calibration curve. The dye removal efficiency (%) was expressed as the percentage ratio of residual dye concentration to that of the initial one. Fourier Transform Infrared (FT-IR) spectroscopy was performed on a Bruker Tensor 27 spectrometer, Germany. The pH was measured by pH meter (654 pH meter Metrohm, Switzerland).

2.3. Batch biological treatment operation

The decolorization experiments were performed in the Erlenmeyer flasks containing 250 ml of the synthetic dye solution and algal biomass under controlled temperature environment in the incubator (Sanyo, Ogawa Seiki Co., Japan). To evaluate the effects of operation and environmental factors on the efficiency of color removal, the batch decolorization experiments were carried out at different initial dye concentrations (2.5–17.5 mg/L), amounts of alga (0.5–6 g), temperatures (5–45 °C) and pH values (1.5–8.5). The pH was adjusted using diluted NaOH and H_2SO_4 solutions.

To examine the reusability of *Vaucheria* sp., repeated-batch operations were used to remove MG. Indeed, 4 g *Vaucheria* sp. was used to decolorize a solution containing 10 mg/L MG. When the decolorization process was completed, the same alga was used to