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Sustainable removal of formaldehyde using controllable water hyacinth

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ABSTRACT

This paper verified the feasibility of water hyacinth for the treatment of formaldehyde in wastewater. In laboratory study, water hyacinth exhibited high removal efficiency for input 100–300 ppm formaldehyde. Formaldehyde consumed by water hyacinth root was lifted from the root to stem and then to leaf, and finally it was converted into some acidoid. Particularly, the removal activity was strengthened with enhanced root activity and chlorophyll in water hyacinth in the presence of 0.5 ppm Eupatorium odoratum L. extract. Meanwhile, the growth of water hyacinth was effectively controlled, exhibiting suppressed emergence of sprouts and low growth rate ($\leq$0.032 g.day^{-1}). In pilot study, water hyacinth also exhibited excellent removal ability to formaldehyde during the four recycle experiments and the growth rate was maintained to a low value about 0.023 g.day^{-1}. In conclusion, water hyacinth managed with Eupatorium odoratum L. extract was proposed as a sustainable biotechnical treatment for formaldehyde.

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1. Introduction

Formaldehyde is widely used in various fields such as the manufacture of synthetic resins (Silva et al., 2015) and glue (Cobut et al., 2015), the production of leather (Marsal et al., 2017; Zhang et al., 2017), and the sorption materials for treating wastewater (El-Korashy et al., 2016). These industries usually generate a large number of wastewater containing variable concentrations of formaldehyde, ranging from a few to hundreds of milligrams per liter (Kajitvichyanukul et al., 2008). Formaldehyde seriously damages human health and the environment (Grafstrom et al., 1985). Thus, formaldehyde is classified as a “probable human carcinogen” with LD50 (in rats, oral) as 100 mg kg^{-1} (Dixit et al., 2015).

Technologies used to clean formaldehyde are categorized into three types: physical adsorption using adsorbents of limited capacity and life cycle (Fang et al., 2008), oxidative degradation using catalysts by means of special facility and high energy consumption (Talaiekhozani et al., 2016; Wu, 2014; Yuan et al., 2017) and biological decomposition using various microbes (Amani and Jalilnejad, 2017; Lu et al., 2012). In general, biological treatment is the most commonly method for the removal of formaldehyde. However, the concentration of formaldehyde in effluents is always beyond the endurance capacity of microbes, which limits the direct use of biological treatment methods (Cui et al., 2016). Therefore, development of an energy efficient, operation convenient and sustainable biotechnical treatment process is essential for the management of formaldehyde in wastewater.

Water hyacinth (Latin name as *Eichhornia crassipes*) is a free floating macrophyte having hairy roots. It can grow easily in polluted wastewater and its pH tolerance is estimated at 5.0–7.5 (https://en.wikipedia.org/wiki/Eichhornia_crassipes). Thus it is considered to be a potential green phytoremediation method for the removal of pollutants presented in wastewater (Rezania et al., 2015; Saleh, 2016) such as chromium (Sarkar et al., 2017), Cd (Zhang et al., 2015), arsenic (Alvarado et al., 2008), zinc (Hardy and Raber, 1985), phosphate (Cai et al., 2017), dyes (Guerrero-Coronilla et al., 2015) and radionuclides (Saleh, 2014, 2012). As reported, the growth of water hyacinth would be inhibited by the pollutants like heavy metals, which in turn destroys the effectiveness of phytoremediation ability of water hyacinth. For example, the toxic effect on water hyacinth plant was observed in the presence of Cr(VI) and phenol (Gupta and Balomajumder, 2015).

This study investigated the sustainable phytoremediation ability of water hyacinth to formaldehyde in the presence of Eupatorium odoratum L. (Latin name as *Chromolaena odorata L.*) extract in both laboratory and pilot scale. The removal percentage of formaldehyde in wastewater and its accumulation of formaldehyde in root and

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aerial part (i.e. stem and leaf) of water hyacinth were measured by UV–Vis Spectrophotometry. Next, the variations of pH for wastewater, water hyacinth root juice and aerial part juice were determined with a pH meter. Finally, the growth of water hyacinth was estimated by means of fresh weight, emergence of sprouts, root activity and chlorophyll content in aerial part.

2. Materials and methods

2.1. Materials

2.1.1. Chemicals

Formaldehyde was purchased from Tianjin Kemiou Chemical Reagent Co., Ltd. Eupatorium odoratum L. extract was prepared in the laboratory as described in our invention patent (patent no. ZL201510485847.8). All of the solvents and chemicals used in the experiment were of analytical grade and used without further purification.

2.1.2. Plants

Water hyacinth was collected from Dianchi Lake situated in the south of Kunming city, China. Homogenous water hyacinth plants at seedling stage of similar fresh weight were selected for the experiment.

2.2. Methods

2.2.1. Removal process in the laboratory

In this study, the removal of formaldehyde was investigated through two different experiments. In the first experiment, an aliquot of 10 L solution containing formaldehyde (concentrations as 0, 100 ppm, 200 ppm and 300 ppm) was prepared and poured into a plastic tank ($48 \times 36 \times 20\ cm^3$) and three replicates were carried out at every formaldehyde concentration level. Ten water hyacinth plants of fresh weight 70 ± 5 g were put into each tank at the beginning of experiment and about 50% water surface area was covered by aerial part of these plants. The original mass ratio of water hyacinth and wastewater was about 0.6:10. The removal experiments were conducted at 10 °C, 20 °C, 30 °C and 40 °C for a period of 10 days. The formaldehyde content in wastewater was analyzed every two days with an UV–Vis spectrophotometer at the maximum absorption wavelength of the derivatization of formaldehyde with acetylacetone (414 nm). The removal percentage of formaldehyde was calculated by the following equation (1):

$$\text{Removal of formaldehyde}\ (\%) = (C0 - Cx)/C0 \times 100\% \quad (1)$$

where C0 and Cx standed for formaldehyde concentration before experiment and after x day treatment, x = 2, 4, 6, 8 or 10.

Every two days, the pH of wastewater was determined with a pH meter. The juice of water hyacinth root or aerial part was squeezed out and its pH and formaldehyde content were analyzed.

In the second experiment, Eupatorium odoratum L. extract was added into the wastewater to obtain a concentration of 0.5 ppm. Other conditions and operations were the same as the first experiment. Every two days, both the fresh weight and the new sprouts were measured to reveal whether the growth of water hyacinth was controllable or not. The mean relative growth rate (RGR) (You et al., 2014) was calculated by the following equation (2):

$$RGR = (\ln W2 - \ln W1)/t \times 100\% \quad (2)$$

where, $W1$ was defined as the mean initial fresh weight, $W2$ as the fresh weight in the final status and t as the time interval (the experimental period).

In order to elucidate the changes in formaldehyde removal efficiency, the root activity and the chlorophyll content in aerial part were determined to reveal the biological activity of root and aerial part. The root activity was measured with an UV–Vis spectrophotometer at the maximum absorption wavelength of the oxidation of *a-naphthylamine* (520 nm) through the enzymatic pathway of root peroxidases (Chiu and Chou, 2012; Yang et al., 2004) and the chlorophyll content in aerial part was measured with a SPAD502 chlorophyll meter.

2.2.2. Removal process in pilot scale

Outdoor pilot test was carried out in a tank ($10 \times 5 \times 1\ m^3$) with $25\ m^3$ wastewater containing 200 ppm formaldehyde and 0.5 ppm Eupatorium odoratum L. extract. Water hyacinth of average fresh weight 200 ± 20 g was put into the first wastewater tank and these plants uniformly floated on the surface of wastewater and covered 50% water surface area. The original mass ratio of water hyacinth and wastewater was set as 0.6:10. The highest day-time temperature was lower than 30 °C and the lowest night-time temperature was higher than 10 °C in Kunming, China. The formaldehyde concentrations in wastewater at 36 sites (Fig. 1) were determined every 12 h until it became to zero. At the end of removal process, formaldehyde accumulated in root and aerial part of water hyacinth was determined. Next, water hyacinth was carefully collected and directly recycled to treat the second tank of wastewater. The treatment conditions were the same as the first removal trial. In this study, water hyacinth was proposed to be recycled four times and five tanks of wastewater were treated in total.

3. Results and discussions

3.1. Removal of formaldehyde in the laboratory

3.1.1. Removal efficiency

Formaldehyde could be effectively removed with water hyacinth and the removal percentage was directly correlated to the temperature and the input of formaldehyde (Fig. 2). At low temperature (10 °C) or high temperature (40 °C), the removal percentage was low. The most effective treatment for formaldehyde was found at 20 °C. After 8-day treatment, all formaldehyde was removed at 20 °C and 100 ppm input. When the input was elevated to 200 ppm, about 92.7% of formaldehyde was removed in 10 days at 20 °C, while less than 88% of formaldehyde could be removed at other temperatures. At the same conditions, the removal percentage was lower with higher input of formaldehyde. For example, at 30 °C, the final removal percentage for formaldehyde reached to 100% for 100 ppm input, while it dropped to a value of 88.7% for 200 ppm input and 40% for 300 ppm input. These results demonstrate that the removal of formaldehyde should be further

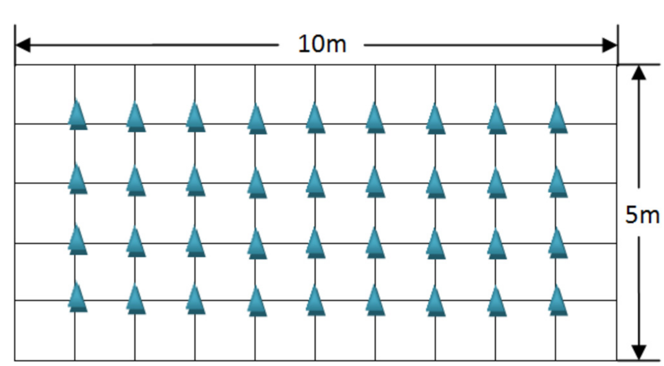

Fig. 1. Diagram for sampling of 36 wastewater samples in each tank.

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