

# Biological decomposition of herbicides (EPTC) by activated sludge in a slurry bioreactor

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## Abstract

Biological treatment of hazardous chemical substances emitted into the environment has been expected to be a desirable treatment method. In this study the biodegradation of an herbicide, *S*-ethyl dipropylthiocarbamate (EPTC), has been investigated in an airlift bioreactor in batch experiments using waste activated sludge, freely suspended and immobilized, under aerobic conditions. The influence of activated sludge loadings (MLSS = 2000–9000 ppm), EPTC concentrations ( $S_0 = 2$ –11 ppm) and aeration rates ( $Q_g = 0.4$ – $3.5 \text{ l min}^{-1}$ ) on the degradation rate was discussed. The results show that for a constant activated sludge concentration of 4162 ppm, the biological degradation rate of EPTC increased with increasing aeration rate up to  $2.5 \text{ l min}^{-1}$ . Although the increase of activated sludge concentration enhanced the biological degradation rate, the higher aeration rate was required to suspend activated sludge uniformly through the reactor and supply enough oxygen to the activated sludge. The maximum biodegradation rate was  $0.16 \text{ h}^{-1}$  at an activated sludge loading of 6000 ppm and aeration rate of  $3.5 \text{ l min}^{-1}$ . Also, it was observed that the biodegradation rates for immobilized activated sludge were higher than those for freely suspended activated sludge. Finally a mathematical model was proposed to describe the biodegradation of EPTC in the bioreactor.

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## 1. Introduction

Herbicides are widely used to control unwanted plant species that are competing for light, water, and nutrients with the wanted plant species. Whenever herbicides are applied, they are inevitably transported to areas distant from the original site of application, by means of rain-induced run-off, wind or rain erosion of soil particles containing traces of herbicide, and evaporation into the atmosphere and subsequent transportation in fog and clouds. Whatever the method of transport, much of the herbicides we apply end up in our rivers and streams. Many herbicides and their additives are directly toxic to aquatic invertebrates and fish. Herbicides kill algae and aquatic plants in waterways which can lead to nitrification and eutrophication and destroy the food base for other aquatic organisms at the base level. Biological degradation of herbicides for the most part involves recognition of the herbicide by one or more

enzymes. Complete mineralization of herbicide to carbon dioxide may require the cooperation of more than one type of organism, waste activated sludge as one example of mixed cultures can be utilized for degrading herbicides. Activated sludge is well known biomass used for the purification of some industrial effluents and domestic wastes [1]. Many reactions involved in pesticide degradation are carried out more rapidly by aerobic organisms, and some reactions actually require molecular oxygen as a substrate. One of the more common steps in pesticide degradation by aerobic organisms is the manner in which aromatic rings are decomposed. Microbes tend to activate aromatic rings prior to ring cleavage by introducing hydroxyl groups.

Bioremediation is the most attractive method for environmental protection due to its cost effectiveness and is unique by offering the potential for complete destruction [2,3]. Therefore, there is a great need for new, highly effective bioprocesses for degradation of recalcitrant pollutants dissolved in water. Out of a range of bioremediation technologies, treatment in a slurry bioreactor is considered to be one of the fastest bioremediation methods since substrates can be effectively transported to the

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microbial population [4]. Among the slurry bioreactors used, airlift systems are a popular choice for many bioreaction processes. Ease of construction and low cost of operation have made them a popular choice for research with suspended cells. Very few literatures are available concerning the biodegradation of herbicides by activated sludge. Zhang et al. [1] developed a novel kind of internal airlift loop bioreactor with cells immobilized onto ceramic honeycomb support installed in a draught tube to overcome some of the drawbacks involved in the treatment of the organic wastewater by bioreactors with immobilized cells including packed bed, fluidized bed or airlift bioreactors. Harding et al. [5] used external loop airlift bioreactor for bioremediation of toluene contaminated air using submerged culture rather than packing materials which can plug during biofilter operation. Recently a new modification of an airlift reactor has been proposed [6]. The draft tube of this reactor is porous and semipermeable and made of non-woven geotextile. It has been found that the liquid flow structure in this reactor is quite unusual compared to classic airlifts. This flow structure is appropriate for the process of attaching soil particles to a static support. The first application of this bioreactor for pentachlorophenol mineralization was very promising [7]. Visvanathana et al. [8], investigate the biodegradation of herbicides (pentachlorophenol) to treat wastewater contaminated with PCP and NaPCP in order to meet the effluent discharge standards, using acclimatized activated sludge in a membrane bioreactor (MBR). Mangat and Elefsiniotis [9] investigated the biodegradation of the herbicide, 2,4-dichlorophenoxyacetic acid (2,4-D) in sequencing batch reactors, the study was conducted to investigate the effects of hydraulic retention time (HRT), the presence or absence of supplemental substrate and variation in feed concentration on the biodegradation potential of 2,4-dichlorophenoxyacetic acid (2,4-D). Hill et al. [10], investigated the potential of the activated sludge treatment process for the removal of chlorophenoxy herbicides (CPH) from domestic waste water using a series of laboratory jar tests and pilot plant experiments. To our knowledge, no studies on the degradation of EPTC herbicides from wastewater employing immobilized riser in an internal loop bioreactor have been described. Therefore, in this study, an attempt has been made to investigate the biodegradation kinetics of a herbicide, *S*-ethyl dipropylthiocarbamate (EPTC), using waste activated sludge (freely suspended and immobilized) in an internal loop airlift slurry bioreactor. Modeling of biodegradation kinetics has been also carried out using a simple modified Monod model to consider substrate inhibition kinetics and compared with experimental results.

## 2. Materials and methods

A schematic diagram of the airlift bioreactor used for batch biodegradation experiments in this work is shown in Fig. 1. The airlift bioreactor was constructed from plexiglass cylinder with an internal diameter of 96 mm and 150 cm height and concentric draft tube of 70 cm internal diameter and 110 cm height. The draft tube was inserted in the reactor in order to enhance the circulation of the activated sludge. Air entered at the base through a glass filter gas sparger. The airflow rate was measured by a calibrated rotameter.

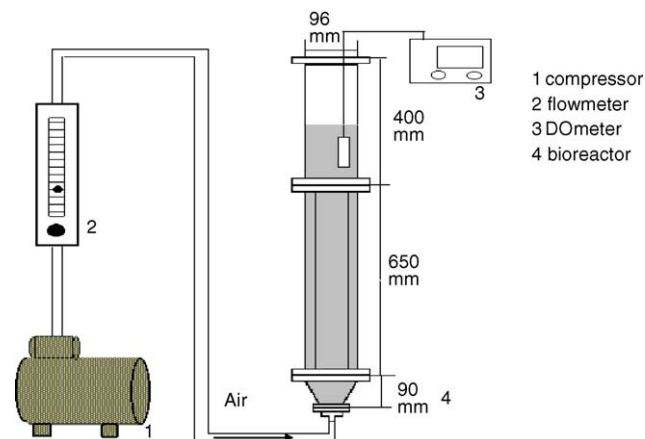


Fig. 1. The experimental set up of the slurry airlift bioreactor.

Experiments were carried out with different initial EPTC concentrations ( $S_0 = 2, 3, 4, 5, 6$  and  $11$  ppm), aeration rates ( $Q_g = 0.4\text{--}3.5$  l min<sup>-1</sup>) and activated sludge concentrations (MLSS = 2000, 4000, 5000, 6000, 7000, 8000 and 9000 ppm).

A waste activated sludge obtained from Kawagoe City Wastewater Treatment Plant located in Saitama, Japan was used as a mixed culture after acclimation.

The activated sludge was acclimatized for three weeks, starting with 10 ppm EPTC solution and slowly raised to 20 ppm under aeration at room temperature and the composition of the nutrient solution used was:  $K_2HPO_3$  217 mg l<sup>-1</sup>,  $KH_2PO_3$  85 mg l<sup>-1</sup>,  $Na_2HPO_3$  450 mg l<sup>-1</sup>,  $NH_4Cl$  17 mg l<sup>-1</sup>,  $MgSO_4$  225 mg l<sup>-1</sup>,  $CaCl_2$  275 mg l<sup>-1</sup>,  $FeCl_3$  2.5 mg l<sup>-1</sup>,  $NaNO_3$  100 mg l<sup>-1</sup>.

For the immobilized activated sludge experiments. Acclimated activated sludge (MLSS = 2136 mg l<sup>-1</sup>) was rapped with a non-woven polypropylene textile with a mesh size of 26. Then the draft tube was filled with the immobilized activated sludge.

Samples (20 ml) of the wastewater were taken from the bioreactor in a regular time for analyzing the herbicides (EPTC) and the mixed liquor suspended solids (MLSS) concentrations. Samples were centrifuged at 2500 rpm to remove the solids from the solutions. To measure the EPTC concentration, after filtering the liquid phase, 10  $\mu$ l was injected in the gas chromatography (SHIMADZU GC-18A, Japan) with a Zebtron ZB-1 column (100% polydimethyl siloxane) and flame ionization detector. Helium was used as the carrier gas. The temperatures of the injection port, oven and detection port were 300, 300, and 60 °C, respectively. The MLSS was determined by the standard method of Japanese Sewage Works Association [11]. The solids were dried and then weighted. Since the biomass concentrations changed slowly with time (10 h), it was assumed to be time independent during the biodegradation operation, as well as the results of Karamanev et al. [12] and Maeda et al. [13]. The temperature of all experiments was constant at (25 °C), and the pH was maintained at 7.0 by addition of 1N NaOH.

## 3. Results and discussion

### 3.1. Biodegradation kinetics of EPTC

The typical experimental results at different initial EPTC concentrations at  $Q_g = 3.5$  l min<sup>-1</sup> and MLSS = 5500 ppm are shown in Fig. 2. Even the biodegradation of EPTC is not complete; it is clear that increasing the initial EPTC concentrations resulted in slow depletion. The biodegradation efficiency decreased from 65% at 2 ppm EPTC concentration to 42% at 11.11 ppm. This is can be attributed to the fact that high substrate concentration inhibits the biomass growth and consequently reducing the biodegradation rate.

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