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Biodegradation of acid orange 7 in an anaerobic–aerobic sequential treatment system

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ABSTRACT

The decolorization of azo dye Acid Orange 7 (AO7) was studied in a sequential anaerobic/aerobic reactor system; the anaerobic stage was carried out in a continuous upflow stirred packed-bed reactor (USPBR) filled with biological sludge carbonaceous material (BSCM) and aerobic stage took place in an aerobic membrane bioreactor (aerobic MBR). In a continuous USPBR-BSCM system, azo dye bioconversion rates were about 99% at very short space times (τ) 1.02–1.6 min. In the sequential aerobic stage, hydraulic retention times (HRTs) were determined on removal efficiencies of chemical oxygen demand (COD), resulting about 66% at 48 h. Total organic carbon (TOC) assay showed a removal efficiency about 54.37%, suggesting the degradation of the aromatic amines produced in the anaerobic reactor. Total aromatic amine (TAA) values were 45.30 and 8.97 mg L^{-1} in USPBR and aerobic MBR respectively, resulting in about 80.28% removal efficiency. These results indicate that anaerobic sequential USPBR/aerobic MBR reactor system seems to be an effective and promising system for complete azo dye biodegradation.

1. Introduction

Azo dyes are chemical substances commonly used in textile, pharmaceutical, and food industries and characterized by the N=N bond. Their production is more than 1 million tons per year in the world, and during dying processes, about 40% of this huge amount of azo dyes ends up in wastewaters [1]. There is no adequate process to treat these wastewaters at high concentrations and at soft conditions on the industrial scale for the time being, and the release of these compounds into the environment presents serious problems of pollution related to both aesthetic reasons and their toxicity.

Several methods have been found to treat azo dye wastewaters [2]. Removal techniques for dyes include coagulation, advanced oxidation processes, membrane processes, and adsorption. Among all of the existing techniques, the most economic and environmentally friendly are biological treatments. Because of the fact that azo dyes are artificial compounds and especially designed to be resistant in the natural environment, their biological degradation has serious obstacles. Investigations into the biodegradability of water-soluble azo dyes by an activated sludge process have indicated that, in most cases, these dyes could not be degraded under aerobic conditions. On the other hand, azo reduction can be relatively easy achieved under anaerobic conditions [3]. Moreover, most of the products created by breaking of the N=N bond could be successfully degraded under aerobic conditions. These suggest a sequential anaerobic–aerobic process as the reasonable scheme for treating wastewaters containing azo dyes [4]. In this process the azo dye containing wastewater is first subjected to an anaerobic biological treatment which results in the reduction, and the consequent decolorization, of the azo dye. Reduction of the non-dye organic load also occurs, to some extent, under anaerobic conditions. However, they can be potentially biodegraded aerobically [5]. Therefore, the major role of the aerobic stage is the reduction of both the non-dye and dye organic loading of the wastewater lower to the limits established by the environmental standards.

Previously, we have investigated removal efficient of azo dye Acid Orange 7 (AO7) using a continuous upflow stirred packed-bed reactor (USPBR) filled with biological sludge carbonaceous material (BSCM) was investigated. The BSCM process utilizes granular carbonaceous materials (GCM) as activated carbon to adsorb organics and to act as a support for microorganisms (biofilm), thus facilitating the elimination of organic water

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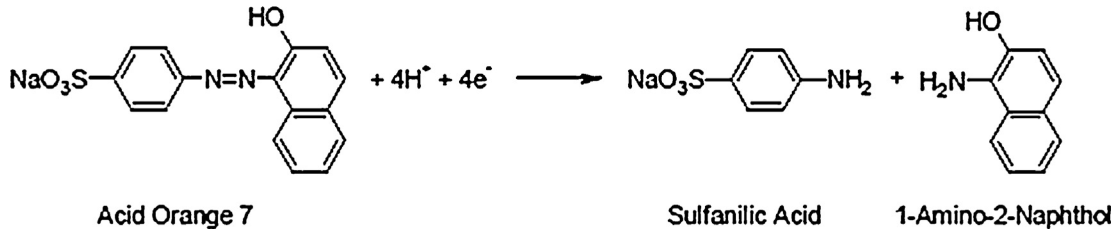

Fig. 1. Anaerobic degradation of Acid Orange 7.

pollutants [6,7]. This study reported that the anaerobic, catalytic reductive azo-dye degradation technique, using BSCM, is an effective and promising treatment for the effective azo reduction [7].

The aerobic membrane bioreactor (aerobic MBR) process, which consists of an activated sludge bioreactor and a microfiltration membrane, is an emerging bio-treatment technology that has demonstrated great promise. It takes advantage of the rapid development in membrane manufacturing and has the potential to fundamentally advance the biological treatment process. Possessing advantages, such as excellent effluent quality, a high biomass concentration without concern for sludge settling problems, a simple flow configuration and small footprint demand encouraged the application of the MBR [8].

Reduction of AO7 to sulfanilic acid (SA) and 1-amino-2-naphthol (1A2N) (Fig. 1) by sewage effluent under anaerobic conditions has been reported in the literature but no a subsequent finishing biological treatment using an aerobic MBR process [9–11]. Based on this observation the present research describes a sequential anaerobic-aerobic process for complete mineralization of AO7. In addition, the influence of periodical stirring of BSCM on decolorization rates in the anaerobic system working in continuous is investigated. The effect of HRT on COD, color and aromatic removals are determined through increasing HRT in the aerobic stage. Finally, total aromatic amine removals in anaerobic/aerobic sequential system in an operation period are studied.

2. Materials and methods

2.1. Chemicals

Azo dye Orange II (C.I. Acid Orange 7, dye content 99%), sulfanilic acid (min 99%), acetic acid (99.8%) and sodium acetate (99%) were purchased from Sigma–Aldrich Company. The sewage sludge from municipal wastewater treatment plant (WWTP) was used to prepare the sludge carbonaceous material (SCM) [7] and get the mixture culture. SCM was crushed and granules of 25–50 mesh size were separated. Activated carbon (Merck, granules of 1.5 mm, ref. 1025141000) was crushed and sieved into 100–200 μm in size. Both were washed with distilled water, dried at 104 °C for 15 h and stored under normal conditions. Carborundum granules (Carlo Erba Reagents, ref. 434766) were used as inert diluent for SCM. The basal media contained the following compounds (mg L^{-1}): $MnSO_4{\cdot}H_2O$ (0.155); $CuSO_4{\cdot}5H_2O$ (0.285); $ZnSO_4{\cdot}7H_2O$ (0.46); $CoCl_2{\cdot}6H_2O$ (0.26); $(NH_4)_6Mo_7O_{24}$ (0.285); $MgSO_4{\cdot}7H_2O$ (15.2); $CaCl_2$ (13.48); $FeCl_3{\cdot}6H_2O$ (29.06); NH_4Cl (190.9); KH_2PO_4 (8.5); $Na_2HPO_4{\cdot}2H_2O$ (33.4); and K_2HPO_4 (21.75). These chemicals were obtained from Sigma–Aldrich Company.

2.2. Experimental set-up

The laboratory scale system consists of an anaerobic USPBR and an aerobic MBR. Schematic diagrams of the anaerobic and aerobic stages are shown in Figs. 2 and 3 respectively.

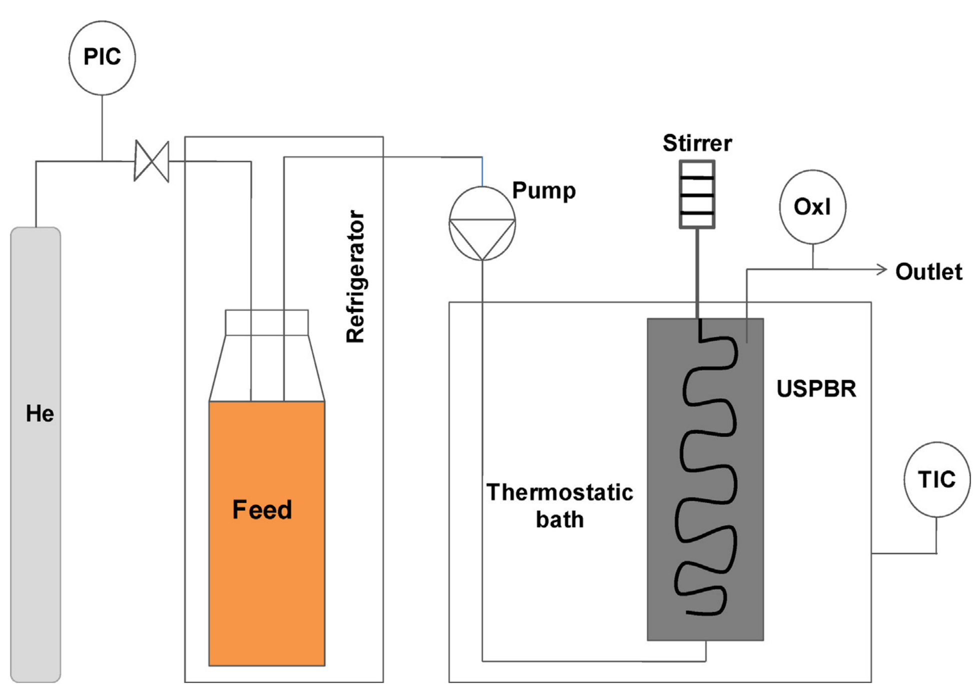

Fig. 2. Anaerobic upflow stirred packed-bed reactor (USPBR) setup.