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Energy-positive nitrogen removal of pharmaceutical wastewater by coupling heterotrophic nitrification and electrotrophic denitrification

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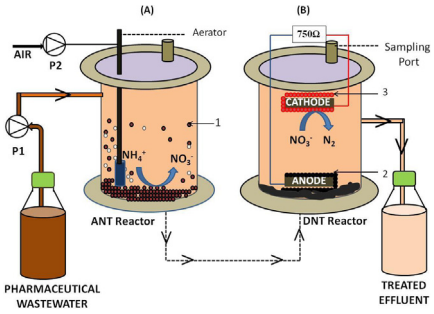
HIGHLIGHTS

- Substantial bioelectrochemical denitrification was noticed under closed circuitry.
- The designed bioelectrochemical DNT system performed at low C/N ratios.
- Sequential integration of ANT & DNT resulted in remarkable multi-pollutant removal.

GRAPHICAL ABSTRACT

Schematic view of (A) ANT (aerobic nitrification treatment) reactor and (B) DNT (denitrification treatment) reactor (1: nitrifying organisms, 2: Anode Oxidizing Bacteria, 3: Nitrate reducing bacteria, P1 & P2: Pumps).



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ABSTRACT

An attempt was made to cohesively assess the aerobic nitrification treatment (ANT) and bioelectrochemical denitrification treatment (DNT) of real-field pharmaceutical wastewater. The ANT resulted in the removal of 73% ammonium and 78.5% organic carbon content. Subsequently, the outlet of ANT reactor was fed to DNT reactor and the relative influence of external circuitry was investigated. The DNT reactor operated with closed circuit mode (CCM) resulted in ~83% of nitrates and % ~61% of organic content removal even at low C/N ratio of 2.18. The other pollutant concentrations *viz.*, phosphates, sulfates and total dissolved salts were also significantly removed by both ANT and DNT processes. In this study, a sequential integration of both ANT and DNT processes exhibited high potential for self-sustained energy-positive nutrient removal.

1. Introduction

In recent years, extensive anthropogenic activities have led to alarmingly over-abundance of nitrogenous compounds (viz., nitrate, nitrite, or ammonium) in water bodies, thus resulting in adverse ecological impact [1]. These compounds like ammonium and nitrate principally together accounts for greater ill-effects on both aquatic as well as a terrestrial ecosystem. In many parts of the world, including India, the potable water is contaminated with nitrate [2]. Therefore, stringent regulations are made mandatory to curtail the concentration of contaminants in industry-discharged water; as in this case ammonium (50 mg L^{-1}) and nitrate (10 mg L^{-1}) [1].

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Conventionally, removal of nitrogenous components from liquid waste is accomplished by employing various physical and chemical methods. However, these methods merely allocate the displacement of ammonium or nitrate from the contaminated solution, instead of destructing these pollutants. Thus, the major shortcoming is the formation secondary wastewater, containing pollutants which is needs to be treated and disposed of safely [3]. Alternatively, the biological approach of nitrification followed by denitrification is found suitable for the removal of ammonium and nitrate [4]. The complete elimination of nitrogen from water through biological route recruits diversified consortia of microorganisms, wherein essential metabolic steps permit the nitrogen remediation process [5]. The ammonium present in wastewater is converted to nitrate during nitrification and further the nitrate content of wastewater is removed by denitrification [6]. The nitrification process is accomplished by nitrifying bacteria in aerobic condition at the optimum pH, where ammonium undergoes stepwise oxidation to form nitrate as the final product [7]. Later, the ubiquitous denitrifying bacteria carry out biological denitrification by utilizing oxygen present in nitrate during their respiratory process in anoxic condition and produce harmless "N_2" by complete reduction [8,9]. In spite of being eco-friendly and economically viable, the sluggish nature of biological approach of nitrogen removal makes it challenging to consider for wastewater treatment [1]. In recent times, efforts have been focused on driving up nitrification and denitrification techniques as it may reduce the complexity of further treatment [10–12]. Recently, bioelectrochemical strategies are gaining prominence for simultaneous wastewater treatment and value-addition [6,13–15].

Earlier studies have documented nitrification and denitrification of synthetic wastewater [9,16]; but the present study aimed to investigate the efficacy of sequential aerobic nitrification treatment (ANT) and bioelectrochemical denitrification treatment (DNT) of real-field pharmaceutical effluent. Furthermore, the influence of external circuitry regulation on DNT reactor performance was investigated.

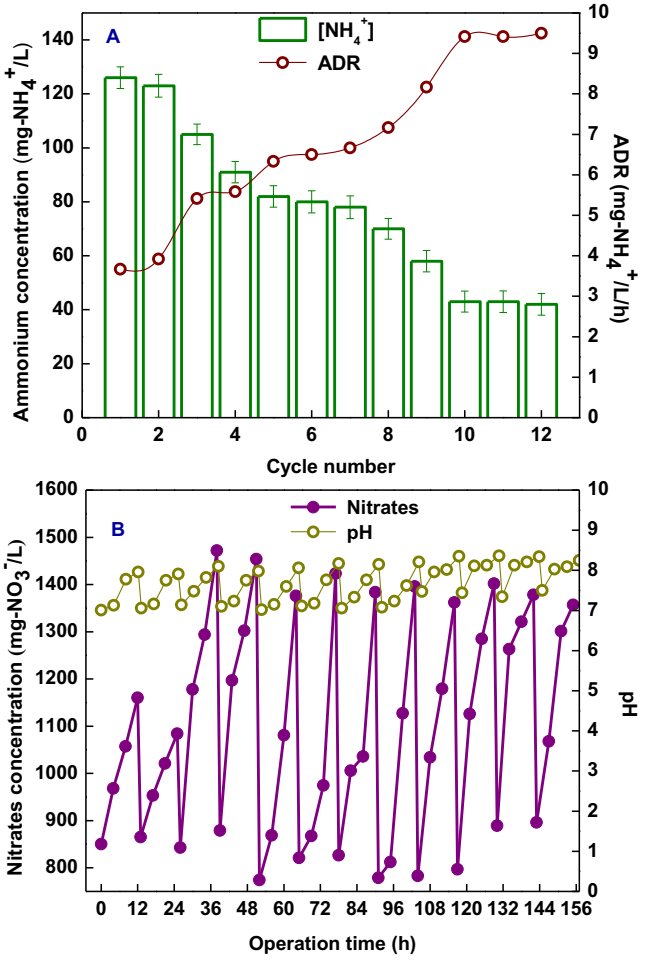

Fig. 1. Nitrification performance of ANT reactor: (A) ammonium removal and ammonium degradation rate (ADR); (B) Variation in nitrate concentration and pH.

2. Experimental methodology

2.1. Biocatalyst

Aerobic mixed culture from aerobic domestic effluent treatment plant was used as inoculum for ANT. Prior to experiment, the aerobic culture was enriched with designed synthetic wastewater (DSW) containing glucose as carbon source and ammonium chloride as nitrogen source. For DNT, anaerobic mixed culture from anaerobic domestic effluent treatment plant was used as inoculum and enriched with DSW containing sodium acetate as carbon source and sodium nitrate as nitrogen source was used for experiment. Thereafter, both the treatment studies were carried out individually with real-field pharmaceutical wastewater which was characterized as follows: pH 6.5–6.7, chemical oxygen demand (COD) – 12,000–13,500 mg L^{-1}, ammonium – 170 mg L^{-1}, nitrate – 985 mg L^{-1}, phosphates – 215 mg L^{-1}, sulfates – 190 mg L^{-1}, TDS – 8450 mg L^{-1}, TSS – 1630 mg L^{-1}, VFA – 2500 mg L^{-1}.

2.2. Reactor construction and operation

Two single-chambered cylindrical reactors were designed and fabricated using perspex material with working/total volume of 1000/1200 mL. Of these, one reactor was used for ANT and the other was used for DNT. The DNT reactor holds a couple of non-catalyzed graphite electrodes (5 cm × 5 cm × 1 cm; geometrical surface area 70 cm^2) as anode and cathode. The anode was completely submerged at the bottom of the reactor that supports exclusively for anaerobic oxidation and cathode at the top that enables anoxic reduction. The entire study is carried out in two phases i.e. ANT followed by DNT. In the first phase, ANT reactor was operated under continuous aeration by maintaining the dissolved oxygen (DO) level of above 0.8 mg L^{-1}. In the second phase, the effluent of ANT reactor was subsequently fed into DNT reactor which was operated in three different circuitry modes *viz.*, open circuit (OCM), closed circuit (CCM) and short circuit (SCM). Initially, the DNT reactor was operated in open circuit mode (OCM) for stabilization of open circuit voltage. Therafter, polarization

Table 1
Relative performance of ANT reactor for ammonium removal.

Cycle number	Inlet ammonium (mg-NH_4^+ l^{-1})	Outlet ammonium (mg-NH_4^+ l^{-1})	Ammonium removal (%)	Outlet nitrate (mg-NO_3^--N l^{-1})	Increase in nitrates (%)	Outlet pH
Cycle 2	165	123	25.45	1024	18.38	7.91
Cycle 8	160	70	56.25	1384	41.89	8.21
Cycle 12	152	42	72.36	1451	64.24	8.41