

Performance of a Two-Stage Ammonia Removal Technology Based on Advanced Oxidation Processes: Effects of Operating Conditions

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Abstract

Ammonia removal and its conversion into recoverable nitrogen species were investigated using a two-stage technology based on advanced oxidation processes. In the first stage, ammonia-contaminated air was treated by UV irradiation, while the second stage used UV/H₂O₂ treatment. The effects of operating conditions on ammonia conversion and the formation of nitrite and nitrate were investigated. The study evaluates the performance of the two-stage process under different operating conditions and examines their role in ammonia conversion and nitrogen product distribution and highlights the importance of optimizing these operating conditions to improve ammonia removal and promote the formation of recoverable nitrogen species.

1 Introduction

Ammonia (NH₃) emissions contribute to air pollution, secondary particulate matter formation, acidification, and eutrophication, with agriculture remaining the dominant emission source in the European Union and the Czech Republic [1–3]. Increasingly stringent environmental regulations therefore require more effective ammonia abatement technologies [1, 4–9].

Available ammonia reduction methods range from preventive measures and improved manure management to end-of-pipe technologies such as biofilters and wet scrubbers. While effective, the latter may involve high operating costs and secondary wastewater generation [3, 10]. Advanced oxidation processes (AOPs), including UV/H₂O₂ and TiO₂-based photocatalysis, offer a promising alternative by enabling ammonia oxidation while producing potentially recoverable oxidized nitrogen species [11–13]. However, most studies have focused on gaseous ammonia removal or dilute aqueous solutions, while the control of nitrite and nitrate formation at higher ammonia concentrations remains less explored [14–16].

Building on our previously developed two-stage technology combining AOPs for ammonia abatement and nitrogen recovery [17], this study investigates the main operating factors affecting its performance. The effects of water vapor in the first UV stage and hydrogen peroxide dosing, initial ammonia concentration, and residence time in the second UV/H₂O₂ stage were examined with respect to ammonia conversion and nitrite/nitrate distribution. The aim was to assess how operating conditions influence ammonia removal and the formation of recoverable oxidized nitrogen species.

2 Material and methods

2.1 Chemicals

Potassium hydroxide were used for pH adjustment. Ammonium hydroxide was used to prepare the ammonia solution for generating ammonia-containing air in the enrichment reactor. Hydrogen peroxide

was used as the oxidizing agent in the chemical scrubber and tubular reactors. All chemicals were of analytical grade and were obtained from commercial suppliers.

2.2 Analytical method

Ammonia concentrations in the gas phase were monitored using an FTIR analyzer (Antaris IGS Analyzer, Nicolet CZ Ltd., Czech Republic). Airflow rate, relative humidity, and temperature were measured using a testo 440 multimeter equipped with appropriate probes (Testo SE & Co. KGaA, Germany). Total nitrogen (TN) in the aqueous phase was determined using a TOC analyzer (FormacsHT-I, Skalar Ltd., Breda, The Netherlands) equipped with an ND25 detector. The pH of the aqueous solution was measured using a Multi 3420 pH meter with a SenTix 940-3 electrode (WTW/Xylem Inc., USA). Nitrite and nitrate concentrations were determined by ion chromatography using an Eco IC system (Metrohm, Czech Republic).

2.3 Experimental apparatus

The experiments were conducted using a two-stage system for ammonia removal and conversion into oxidized nitrogen species. In the first stage, ammonia-containing air was introduced into a stainless-steel photolytic reactor (R1) equipped with low-pressure mercury UV lamps emitting at 185/254 nm. Ammonia-containing air was generated by bubbling air through an ammonium hydroxide solution and subsequently mixed with air before entering R1. The ammonia concentration was monitored by FTIR. The second stage consisted of a wet counter-current scrubber (R2) followed by three tubular reactors (R3) connected in series. The scrubber operated without packing and contained a recirculating hydrogen peroxide solution. Each tubular reactor was equipped with a 254 nm UV lamp positioned inside a quartz tube. The UV/H₂O₂ process generated hydroxyl radicals responsible for the oxidation of ammonia-derived compounds in the aqueous phase. The experimental system was designed and assembled in cooperation with Dekonta Inc.

2.4 Experimental setup

Experiments were performed under controlled ammonia loading and airflow conditions. The experimental conditions were varied according to the objective of each experiment. In a typical experiment, the scrubber was filled with the aqueous solution and liquid circulation was started. Hydrogen peroxide was then added, followed by introduction of ammonia-containing air. After stabilization of the inlet ammonia concentration, UV irradiation was initiated in the first stage and subsequently in the tubular reactors. Aqueous samples were collected at defined reaction times for TN, nitrite, and nitrate analysis. Control samples were collected before and after hydrogen peroxide addition. Selected experiments were performed in batch mode, without gas-phase ammonia introduction and first-stage photolysis. Ammonia conversion in the first stage was calculated from the inlet and outlet gas-phase concentrations. In the second stage, direct ammonia determination was not used for final evaluation because hydrogen peroxide interfered with the Nessler method under the investigated conditions. Instead, total nitrogen together with nitrite and nitrate concentrations was used to evaluate ammonia conversion and nitrogen product formation. Nitrite and nitrate concentrations were expressed

as nitrogen concentrations based on their molar masses. The conversion of ammoniacal nitrogen to oxidized nitrogen species was calculated from the initial and final ammoniacal nitrogen concentrations using the nitrogen balance.

3 Results and discussion

3.1 First stage of technology

The first stage of our technology was based on UV photolysis at 185/254 nm and had been investigated in detail in our previous study [17]. The present work focused on the effect of water vapor on ammonia conversion. Humidification enhanced ammonia removal during UV irradiation, indicating that water vapor contributes to the formation of reactive oxidizing species involved in ammonia oxidation. A certain degree of ammonia removal was also observed without UV irradiation, suggesting that physical uptake into the aqueous phase contributed to the overall removal. The results therefore support controlled humidification as a potential means of improving the performance of the photolytic stage.

3.2 Second stage of technology

The second stage used a UV/H₂O₂ process for the oxidation of ammonia-derived compounds in the aqueous phase. The hydrogen peroxide dosing strategy affected process performance, with more frequent dosing providing more effective oxidant availability. Increasing the initial ammonia loading increased the formation of oxidized nitrogen species but shifted their distribution toward nitrite. This indicates that the subsequent oxidation of nitrite to nitrate becomes increasingly limited at higher ammonia loading. Direct ammonia determination using the Nessler method was found to be unreliable under the investigated conditions due to interference from hydrogen peroxide; therefore, process performance was evaluated using total nitrogen together with nitrite and nitrate concentrations.

4 Conclusion

This study investigated the main operating factors affecting a two-stage ammonia removal technology based on advanced oxidation processes. In the first stage, humidification enhanced ammonia removal during UV irradiation, indicating a beneficial role of water vapor in the formation of reactive oxidizing species.

In the second UV/H₂O₂ stage, the hydrogen peroxide dosing strategy, ammonia loading, and residence time affected both ammonia conversion and the distribution of oxidized nitrogen products. Higher ammonia loading promoted the formation of nitrite, while the subsequent oxidation of nitrite to nitrate became increasingly limited. Increasing residence time alone did not improve process performance, highlighting the importance of hydrodynamic conditions and oxidant availability in addition to irradiation time.

The results provide a basis for further optimization of the two-stage process, with particular emphasis on improving the conversion of nitrite to nitrate and maximizing the formation of recoverable nitrogen species.

Declarations

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