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Evaluation of ammonia–nitrogen removal efficiency from aqueous solutions by ultrasonic irradiation in short sonication periods

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ABSTRACT

In this study, mechanisms and efficiency of ammonia–nitrogen removal from aqueous solutions by ultrasonic irradiation were investigated. Depending on the factors affecting the sonication (initial concentration, initial pH, ultrasonic power density and sonication period), sonication tests were carried out and ammonium–nitrogen removal efficiency by ultrasonic irradiation was determined. In these experiments, ammonia–nitrogen removal efficiency was achieved in the range of 8–64%. In short sonication periods, the best ammonia–nitrogen removal efficiency was achieved at pH 8.2–11. Lower ammonia–nitrogen removal efficiency was observed in high initial ammonia–nitrogen concentration of solutions. It was observed that high initial ammonia–nitrogen concentrations may led to decreased ammonia–nitrogen removal efficiency however quantity of ammonia–nitrogen removal was higher. Because high initial concentration had a negative impact on the sonochemical reactions the heat of cavitation bubbles was reduced. Ammonia–nitrogen removal efficiency was increased with ultrasonic density and sonication period. This study showed that effective ammonia–nitrogen removal could be achieved by the ultrasonic irradiation in short sonication periods (as 60–600 s). Specific cost of ammonia–nitrogen removal by the ultrasonic irradiation from simulated ground water, surface water, wastewater and landfill leachate was also calculated. The specific removal cost was varied between 0.01 and 0.25 $/g ammonia–nitrogen.

1. Introduction

Water is an indispensable necessity on the earth for the continuation of life. However, the limited water resources of the world are being polluted each passing day by the anthropogenic factors that are increasing with human population and industrialization [1]. High ammonia–nitrogen contents and inadequately treated domestic or industrial wastewaters when discharged to receiving water bodies eutrophication and deterioration of water quality problems can be occurred [2]. Besides, rain water flowing from over-fertilized agricultural land is causing environmental problems in the aquatic ecosystem [3]. Ammonia–nitrogen is responsible for unwanted aquatic growth and it is toxic for fishes in the aquatic ecosystem [4]. Furthermore, reduction of ammonia–nitrogen to nitrate by the biological process (nitrification) is causing increasing oxygen consumption in water bodies. Thus, over 10 mg/l nitrate concentration in the water is toxic for human health [5]. Ammonia–nitrogen content of water increased chloride consumption and formation of carcinogenic chlorinated amines in the disinfection process. Moreover, ammonia–nitrogen content of drinking water may cause growth of bacteria in drinking water grid [4]. Today, several physicochemical and biochemical treatment techniques have been used for ammonia–nitrogen removal from domestic and industrial wastewaters [3]. The most commonly used treatment techniques are advanced biological treatment [6], chemical precipitation [7], supercritical oxidation [8], air stripping [9], microwave radiation [10], ion exchange [11], breakpoint chlorination, membrane filtration [12], and adsorption [13].

Today, ultrasound technologies have been commonly used in several fields (textile, medicine, energy production sector, oil–gas industry, food industry, water disinfection process etc.) and water, wastewater and sludge treatment by ultrasound irradiation is one of the most important issues for researchers in the last 15–20 year period [14,15]. Sonication has been used as an advanced oxidation process (AOP) in wastewater treatment [16,17]. The mechanism of sonication is based on sonochemical reactions which are pyrolytic reactions and cavitation. Pyrolytic reactions are realized of high temperature (2000–5000 K) and pressure (500–10,000 atm) inside the formed cavitation bubbles by the ultrasonic irradiation. In this way, free radicals are formed in a very short period (millisecond) such as H, OH, and OH_2 in solution. These radicals are involved

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in the rapid oxidation of organic and inorganic matters and degradation of complex compounds in solution [18,19]. Moreover, toxic compounds turned out to be more easily degradable compounds [18]. Sonochemical reactions can be explained with four main theories which are hot-spot, plasma discharge, electrical and supercritical theories [15]. However, hot-spot theory is the most commonly used theory to explain the sonochemical reactions [20]. According to the hot-spot theory, sonochemical reactions occur in three different zones in homogenous liquid which is inside the cavitation bubbles, gas–liquid interface and bulk solutions (Fig. 1).

- *Inside of cavitation bubbles:* Water is pyrolized to form free radicals such as H, OH, and OH_2 inside the cavitation bubbles by ultrasonic irradiation with the effects of high temperature (2000–5000 K) and pressure (>500 atm).
- *Gas–liquid interface:* The temperature is lower than inside cavitation bubbles between the cavitation bubbles and bulk solution interface. In this zone, oxidative reactions occurred with the effects of pyrolytic and free radicals.
- *Bulk solution:* The free radicals are transferred to bulk solution from the bubbles and interface. Sonochemical oxidation reactions occur between organic–inorganic compounds and free radicals [15].

In this study, solutions with different initial pH and ammonia–nitrogen concentrations were prepared. Ammonia–nitrogen removal efficiencies were determined by the application of ultrasonic irradiation with various (150–735 W) ultrasonic power densities and sonication periods (60–600 s). Thus, different experimental conditions, the effect on ammonia–nitrogen removal efficiency and the removal mechanism were investigated. Ammonia–nitrogen removal costs were also calculated. The main aim of the study was to evaluate the ammonia–nitrogen removal from the aquatic solutions with ultrasonic radiation, even at short sonication periods.

2. Materials and methods

2.1. Materials and chemicals

Ammonium chloride (NH_4Cl) was dissolved in distilled-deionized water and ammonia–nitrogen solutions having various initial concentrations (such as 5.6, 41.5, 84.5, 210 and 415 mg/l) were prepared. Besides, ammonium–nitrogen solutions having different initial pH values (such as 3, 7, 8.5 and 10) and 30 mg/l initial concentration solutions were prepared. The solutions were diluted to initial concentration and adjusted to the required pH with hydrochloric acid and sodium hydroxide.

2.2. Experimental set-up

The sonication tests were carried out at batch test cycles by a laboratory-size sonic device (SONICS VCX 750 Model, 750 W, 20 kHz). The power input could be adjusted continuously from 0 to 750 W. The temperature inside the surrounding bath was maintained by continuous circulation of cooling water. The sonic probe was immersed into the liquid shallow depth and then sonication was started. 250 ml of prepared ammonia–nitrogen solution was sonicated in a covered cylindrical glass vessel. The sonication tests were carried out at 25 ± 2 °C constant temperature. The details of experimental set-up are shown in Fig. 2.

2.3. Experimental procedure

Ammonia–nitrogen solutions having different initial concentrations were prepared. pH of the solutions was adjusted to predetermined pH values before sonication. Ultrasound device was set to the predetermined test condition and then 250 ml solution was sonicated. Sonication test was carried out between 60 and 600 s. After the sonication ammonia–nitrogen contents of the sonicated solution was analyzed with the colorimetric method according to Standard Methods (Nessler Method) [22]. Ammonia–nitrogen analysis was performed with a Palintest 1000 photometer and pH values of solutions were determined with a WTW 330i pH meter.

3. Results and discussions

Initial concentration and pH, ultrasonic power density and sonication period affected the ammonia–nitrogen removal efficiency and sonochemical reactions [20]. Thus, ammonia–nitrogen removal efficiencies and mechanism from the solutions by the ultrasonic irradiation were tested under various experimental conditions.

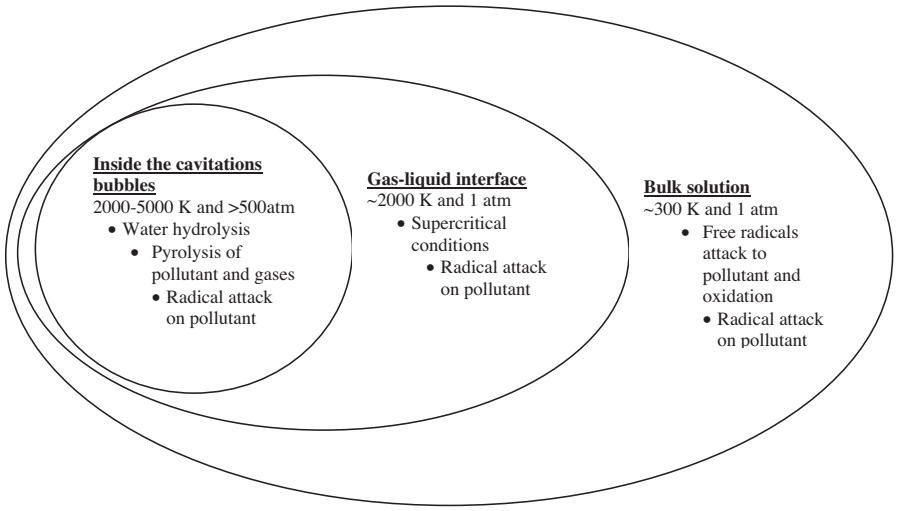

Fig. 1. Reaction zones in cavitation process [21].