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Decontamination of unsymmetrical dimethylhydrazine waste water by hydrodynamic cavitation-induced advanced Fenton process

Mahmood Torabi Angaji [*], Reza Ghiaee

School of Chemical Engineering, College of Engineering, University of Tehran, Tehran, Iran

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ABSTRACT

A pilot scale hydrodynamic cavitation (HC) reactor, using iron metal blades, as the heterogeneous catalyst, with no external source of H_2O_2 was developed for catalytic decontamination of unsymmetrical dimethylhydrazine (UDMH) waste water. In situ generation of Fenton reagents suggested an induced advanced Fenton process (IAFP) to explain the enhancing effect of the used catalyst in the HC process. The effects of the applied catalyst, pH of the initial solution (1.0–9.7), initial UDMH concentration (2–15 mg/l), inlet pressure (5.5–7.8 bar), and downstream pressure (2–6 bar), have been investigated. The results showed that the highest cavitation yield can be obtained at pH 3 and initial UDMH concentration of 10 mg/l. Also, an increase in the inlet pressure would lead to an increase in the extent of UDMH degradation. In addition, the optimum value of 3 bar was determined for the downstream pressure that resulted to 98.6% degradation of UDMH after 120 min of processing time. Neither n-nitrosodimethylamine (NDMA) nor any other toxic byproduct (/end-product) was observed in the investigated samples. Formic acid and acetic acid, as well as nitromethane, were identified as oxidation by-products. The present work has conclusively established that hydrodynamic cavitation in combination with Fenton's chemistry can be effectively used for the degradation of UDMH.

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1. Introduction

UDMH is primarily used as a high-energy rocket propellant. It is also used in manufacturing a plant growth regulator, chemical synthesis, photographic chemicals, as a stabilizer for fuel additives and as an absorbent for acid gases [1]. UDMH is known as a primary eco-toxicant with the maximum permissible concentration in ambient water as low as 0.02 mg/l [2]. Once occurred in the soil or water under natural conditions, UDMH is spontaneously oxidized to form n-nitrosodimethylamine (NDMA). NDMA is a probable human carcinogen with 10^{-6} lifetime cancer risk associated with a drinking water concentration of 0.7 ng/L determined by the U.S. Environmental Protection Agency (USEPA) [3]. It is also recognized as 2A, probably carcinogenic by the International Agency for Research on Cancer of the World Health Organization (IARC) [4].

The traditional technique known for the UDMH stocks abatement is a catalytic incineration [5], which is not appropriate for the treatment of water and air containing UDMH as a pollutant. Oxidation with ozone, catalytic oxidation with oxygen and hydrogen peroxide in the presence of Cu, Fe, Co salts supported on zeolites as catalysts, and oxidation with chloramine may be used for the removal of UDMH from water [6,7]. The main disadvantage of these methods is formation of NDMA as an intermediate of UDMH oxidation [8]. Advanced oxidation processes (AOPs), such as photocatalysis, Fenton, photo-Fenton, and UV/H_2O_2 have been investigated and reported by some researchers as efficient methods for degradation of UDMH, and minimizing the formation of NDMA [9–11]. However, the continuous need for an oxidizing agent (hydrogen peroxide, ozone, etc.) makes these methods inefficient for industrial applications.

Hydrodynamic cavitation (HC) is one form of AOPs. HC has shown considerable promise for wastewater treatment applications due to the ease of operation and its ability to be applied for in situ production of hydroxyl radicals (a strong oxidizing agent). Sivakumar et al. [12] have studied the use of HC reactors for degradation of Rhodamine-B dye solution. Saharan et al. [13] have investigated the use of HC reactors for degradation of orange-G dye using three different cavitating device. On the other hand, there are not many reports indicate individual applications of HC reactor for wastewater treatment, possibly due to the lower intensity of hydroxyl radical generated in the reactors.

The best approach for utilizing the impressive properties of HC is to use it in combination with other AOPs, and exploit the

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[*] Corresponding author. Tel.: +98 2166498982; fax: +98 2166987784.
E-mail addresses: m.t.angaji@gmail.com, mtorabi@ut.ac.ir (M. Torabi Angaji).

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List of acronyms

AOPs	advanced oxidation processes
ED	extent of degradation
HC	hydrodynamic cavitation
IAFP	induced advanced Fenton process
IARC	International Agency for Research on Cancer
IMBs	iron metal blades
NDMA	n-nitrosodimethylamine
UDMH	unsymmetrical dimethylhydrazine
USEPA	United State Environmental Protection Agency

synergistic effects expected due to a common mechanism of destruction. Chakinala et al. [14] have reported the applicability of a combination of HC and chemically-induced advanced Fenton process for treatment of industrial effluents. Bagal et al. [15] have investigated the use of HC reactors in combination with chemical oxidation processes for degradation of 2,4-dinitrophenol. Bagal et al. also have studied the feasibility of coupling a HC reactor with conventional Fenton, and Fenton-like process.

The present work reports the use of HC for inducing advanced Fenton process as a useful chemical-free method for decontamination of waste water. The aim of the work is establishing an efficient treatment strategy for the destruction of UDMH. The outcome of different operating parameters on the extent of degradation (ED) of UDMH has been established while the effectiveness of the treatment performance of the process described has been determined by monitoring the ED at regular time intervals.

The use of zero-valent iron in the form of iron metal blades (IMBs) for intensification of the production of hydroxyl radicals in a similar way to the normal Fenton reaction, using inexpensive and simple techniques for complete removal of UDMH from waste water, and the lack of production of toxic byproducts make this research unique.

2. Materials and methods

2.1. Materials

Unsymmetrical dimethylhydrazine ($C_2H_8N_2$, CAS-Nr.: 57–14-7, 99%), formaldehyde (CAS-Nr.: 82115–62-6, 2%), sulfuric acid (97% Merk), sodium hydroxide (Sigma–Aldrich), hydrochloric acid (37% Merk), FerroVer® iron reagent (Catalog Nr.: 2105769, Hach, 50%), and disodium salt of the chromotropic acid dihydrate (Aldrich) were used in the study. All chemicals were utilized without any further purification. Deionized water was used to prepare all solutions. IMBs were cut from a main single sheet, in desired sizes.

2.2. HC reactor configuration

As it is shown in Fig. 1, the experimental setup is essentially a closed-loop HC reactor that operates in re-circulation mode. The reactor comprises: (1) a holding tank of 200 l volume, (2) a cooling system to adjust and control the temperature, (3) a centrifugal pump (MOTTAHED Industrial and Manufacturing Co., MCP 50-170, 3.0 kW, 2850 rpm.), (4) a multiple-hole orifice plate accommodated with flanges, (5) a catalyst chamber, (6) a Turbine type flowmeter (Hoffer Flow Controls, INC., HO-L-110), (7) data acquisition system, flow control valves (V_1–V_4), and pipes.

The used pipes have an inner diameter of 38 mm and an external diameter of 40 mm. The suction side of the pump is connected to the bottom of the holding tank. The discharge from the pump branches into two lines. The main line consists of an orifice plate which acts as a cavitating device, located just after the upstream piezoelectric pressure gauge (P_2). A bypass line and control valve (V_1) are provided to control the liquid flow through the main line. During the experiment, the bypass valve (V_2) was kept open till the pump reached its maximum speed and then totally or partially closed to achieve the desired pressure in the inlet of cavitation device. The main and bypass lines terminate well inside the tank, below the liquid level, to prevent the induction of gas (air) into the liquid due to the plunging liquid jet. Control valves are provided at appropriate places to manipulate or maintain the flow rate in the main line. Piezoelectric pressure gauges are provided to measure the pump outlet pressure (P_1), the orifice plate inlet pressure (P_2), and the orifice plate downstream pressure and (P_3). Fig. 2 shows a schematic view and the dimensions of the applied orifice plate.

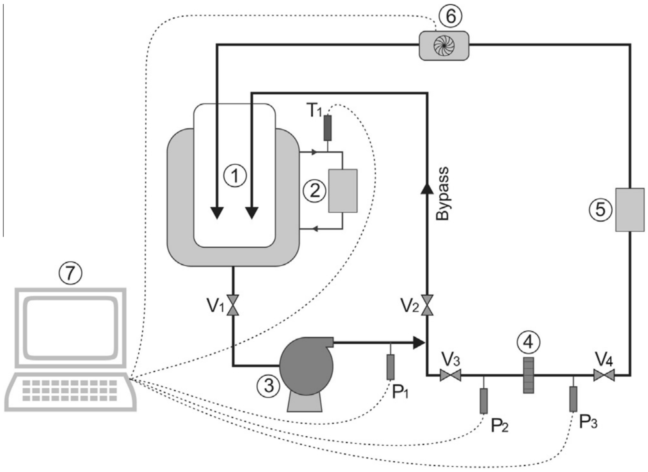

Fig. 1. Schematic diagram of the experimental setup.

Fig. 3 shows the arrangement of the iron blades in the catalyst chamber. The chamber enclosed 16 pieces of IMBs with dimensions of 15 × 60 × 1 mm. IMBs were used after proper washing with no more surface preparation and new IMBs were employed for each test. The orifice plate, catalyst chamber, flanges, holding tank, pipeline, valves and their fitting were made of stainless steel (grade 316L).

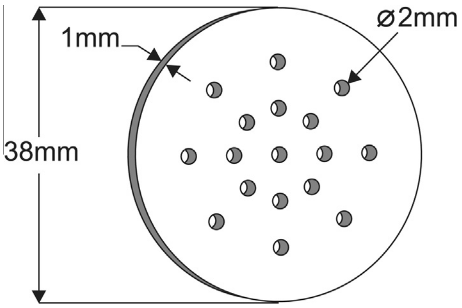

Fig. 2. Schematic view of the applied orifice plate.

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