



Intensification of degradation of methomyl (carbamate group pesticide) by using the combination of ultrasonic cavitation and process intensifying additives



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ABSTRACT

In the present work, the degradation of methomyl has been carried out by using the ultrasound cavitation (US) and its combination with H₂O₂, Fenton and photo-Fenton process. The study of effect of operating pH and ultrasound power density has indicated that maximum extent of degradation of 28.57% could be obtained at the optimal pH of 2.5 and power density of 0.155 W/mL. Application of US in combination with H₂O₂, Fenton and photo-Fenton process has further accelerated the rate of degradation of methomyl with complete degradation of methomyl in 27 min, 18 min and 9 min respectively. Mineralization study has proved that a combination of US and photo-Fenton process is the most effective process with maximum extent of mineralization of 78.8%. Comparison of energy efficiency and cost effectiveness of various processes has indicated that the electrical cost of 79892.34 Rs./m³ for ultrasonic degradation of methomyl has drastically reduced to 2277.00 Rs./m³, 1518.00 Rs./m³ and 807.58 Rs./m³ by using US in combination with H₂O₂, Fenton and photo-Fenton process respectively. The cost analysis has also indicated that the combination of US and photo-Fenton process is the most energy efficient and cost effective process.

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1. Introduction

Methomyl is a broad spectrum insecticide which belongs to the family of oxime carbamate pesticides [1]. It has been widely used all over the world because of its powerful control against many insects and pests. It can easily cause contamination of both ground and surface water resources, due to its high solubility (57.9 g/L, 20 °C) in water and a low-sorption affinity to soils [2]. Presence of methomyl in water bodies can cause potential impact on the human health and environment because of its toxic and hazardous nature [3].

Various studies have been reported in the literature for the degradation of methomyl, which mainly includes the application of advanced oxidation processes such as photocatalytic [2–5] and Fenton, photo-Fenton processes [1,5,6]. Tamimi et al. [1] have investigated the degradation of methomyl in aqueous solutions

by using Fenton (H₂O₂/Fe²⁺) and photo-Fenton (H₂O₂/Fe²⁺/UV) process. Complete degradation of methomyl was achieved after 60 min of operation with 23% and 48% of TOC reduction in case of Fenton and photo-Fenton processes respectively. In other study, Tamimi et al. [2] have reported that photocatalytic degradation of methomyl using Degussa-P25 TiO₂ could lead to 80% TOC removal in less than 4 h. Tomasevic et al. [3] have studied the photocatalytic degradation of methomyl in water using Fe-ZSM-5 zeolite and AlFe-pillared montmorillonite catalysts under halogen lamp light and obtained 100% TOC removal within 4 h using 5 g/L Fe-ZSM-5 zeolite. Malato et al. [5] have also carried out photocatalytic degradation of water-soluble pesticides (diuron, imidacloprid, formetanate and methomyl) by the application of photo-Fenton and solar light driven photocatalytic process using TiO₂. Photo-Fenton process was found to be more efficient than the photocatalysis process on the basis of both, the extent of degradation as well as extent of mineralization of methomyl. Mico et al. [6] have reported enhancement in the performance of photo-Fenton reaction at high salinity conditions when applied for the oxidation of methomyl. However, the degradation of methomyl by the application of ultrasound cavitation is not yet reported in the literature, to the best of

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our knowledge. Ultrasound cavitation is an emerging advanced oxidation process which can be used for the effective degradation of organic pollutants [7]. It consists of mainly three steps occurring repetitively: nucleation, rapid growth of cavities during the compression/rarefaction cycles and the implosion of the cavities once they reach a critical size [8]. Implosion of the cavities subsequently induces thermal dissociation of water and dissolved oxygen molecules to convert them into reactive species such as hydroxyl radicals (OH^\cdot), hydrogen atoms (H^\cdot), oxygen atoms (O^\cdot) and hydroperoxyl radicals (HO_2^\cdot) (Eqs. (1)–(3)) [8,9]. In addition to this, ultrasound cavitation also generates hot spots, strong acoustic streaming and intense fluid turbulence at very short time scale [10].



where “US” signifies ultrasound waves.

The radicals generated may reach the water-cavity interface or diffuse into the bulk solution where they can oxidize the target pollutant. However, the degradation efficiency of ultrasonic reactors is not very promising and economically attractive if applied individually. For this reason, ultrasound cavitation is generally combined with H_2O_2 , Fenton, photo-Fenton, ozone, photocatalytic process, etc. for the intensification of degradation of organic pollutants [7,8].

Present work deals with the application ultrasound cavitation in combination with H_2O_2 , Fenton and photo-Fenton process for accelerating the rate of degradation of methomyl that can be otherwise obtained using individual ultrasound cavitation. It also includes the optimization of operating parameters such as initial pH of the solution and ultrasound power density for maximizing the performance of ultrasound cavitation. In addition to this, extent of mineralization, energy efficiency and cost effectiveness of individual US and hybrid processes have also been studied in order to obtain the most effective and economical process for the degradation of methomyl.

2. Materials and methods

2.1. Materials

Commercial grade methomyl (Dupont) was used as a model organic pollutant without further purification. The structure of the methomyl is as shown in Fig. 1. Hydrogen peroxide (30% w/v), ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), sodium sulfite (Na_2SO_3) and sulfuric acid (H_2SO_4) all of AR grade were obtained from Thomas Baker, Mumbai, India. Acetonitrile and water used for HPLC analysis were purchased from S D Fine Chemicals Ltd., Mumbai, India of HPLC grade. All chemicals were used as received from the supplier.

2.2. Experimental set up

Schematic representation of experimental set up has been given in Fig. 2. It essentially consists of ultrasonic processor, quartz reactor, temperature indicator, UV source and wooden UV shield. Ultrasonic cavitation was induced by using a low frequency (20 kHz) ultrasonic processor (make-Johnson Plastosonic Pvt. Ltd., India) having a tip diameter of 13 mm and maximum power dissipation of 500 W with a facility of varying amplitude viz. 25%, 50%, 75% and 100%. The tip was immersed in the aqueous solution of methomyl to the depth of 1 cm. All the experiments were carried out in a 250 mL quartz reactor with a cooling jacket to control the

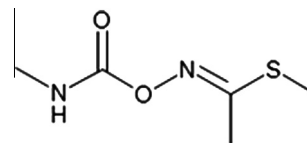


Fig. 1. Molecular structure of methomyl.

temperature increase due to cavitation effects and UV irradiation. The reaction temperature was maintained at $28 \pm 3^\circ\text{C}$ (ambient conditions) by continuously circulating cold water through the cooling jacket. UV source was used in case of photo-Fenton process and its combination with ultrasonic cavitation. Wooden UV shield has been provided to eliminate the possibility of exposure to harmful UV irradiation.

2.3. Experimental methodology

All the experiments were performed with 250 mL aqueous solution of methomyl having initial concentration of 25 ppm. Initially, the effect of power density (i.e. energy input per milliliter of solution) on the rate of sonochemical degradation of methomyl was studied by varying the ultrasonic irradiation amplitude viz. 25%, 50%, 75% and 100%. Calorimetric study was performed to obtain the calorimetric efficiency of the horn at varying operating amplitudes. Further, the effect of initial pH on sonochemical degradation of methomyl was also investigated by varying the pH in the range of 2.5–7.5 (2.5, 3, 5 and 7.5). Initial pH was adjusted by adding H_2SO_4 as per the requirement.

In order to increase the degradation efficiency of methomyl, the experiments were also carried out by combining the ultrasonic cavitation with process intensifying additives such as H_2O_2 and Fenton reagent. The effect of loading of H_2O_2 on sonochemical degradation of methomyl was investigated by varying the molar ratio of methomyl to H_2O_2 viz. 1:10, 1:20, 1:30, 1:40 and 1:50 for fixed initial concentration of methomyl (25 ppm) to determine an optimal concentration of H_2O_2 .

The combined process of ultrasonic cavitation and Fenton chemistry was also employed for the degradation of methomyl. The effect of loading of ferrous ions on sonochemical degradation of methomyl has been evaluated by varying molar ratio of Fe^{2+} : H_2O_2 viz. 1:50, 1:40 and 1:30 at fixed optimal concentration of H_2O_2 . Furthermore, the combined process of ultrasonic cavitation and photo-Fenton process was also utilized to further enhance the rate of degradation of methomyl. Two UV lamps of 8 W each were used as a source of ultraviolet light. In order to obtain the synergistic effect, the efficacy of hybrid processes (combinations of AOP's) was compared with the efficacy of individual processes based on the rate and the extent of degradation of methomyl accomplished. For this reason experiments were also carried out at optimized loadings of additives in the absence of ultrasound i.e. conventional approach of only stirring.

The mineralization study of methomyl was also carried out by analyzing total organic carbon content of the samples obtained at optimized conditions.

2.4. Analysis

High pressure liquid chromatography (HPLC) equipped with C18 column of dimensions 4.6×250 mm (make-JASCO, Japan Spectroscopic Corporation, Japan) was used to determine the concentration of methomyl at a set interval of time. The mobile phase consists of a mixture of 30% acetonitrile and 70% water with flow rate maintained at 1 mL/min and UV detector set at wavelength of 234 nm. Quenching agent (0.5 N sodium sulfite) has been added

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