Science & Technology Development Journal – Science of The Earth & Environment, 5(2):417-423

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Research article

Electron beam induced degradation of atrazine in solution using Taguchi approach

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History

- Received: 17-8-2021
- Accepted: 17-9-2021
- Published: 07-11-2021

DOI : 10.32508/stdjsee.v5i2.653

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ABSTRACT

This study investigated the removal of atrazine from an aqueous solution upon electron beam irradiation from an electron accelerator. Electron beam irradiation could be considered an advanced oxidation process (AOP); these techniques have been recently applied to remove a lot of contaminants in wastewater streams. Atrazine concentrations in aqueous solutions ranging from 2 mg/L to 6 mg/L were eliminated using electron beam irradiation (2-6 kGy) at pH levels ranging from 5 to 9. The coupled electron beam and hydrogen peroxide (from 1 to 5 mM) were also investigated. This study was conducted by the Taguchi method with four variables: initial pH, atrazine concentration, H_2O_2 dosage, and absorbed dose to mitigate atrazine in solutions. The Taguchi process was evaluated using a Signal to Noise (S/N) ratio to find the optimal condition with the simplest design. The obtained results indicate that the absorbed dose is the most important factor, followed by the atrazine concentration and initial pH, while H_2O_2 seems negligible to the removal efficiency. The optimal Taguchi condition shows that the electron beam process reached the best efficiency. The best predicted atrazine eradiation was obtained 100.1% at initial pH 5, H_2O_2 of 3 mM, atrazine concentration of 2 mg/L and absorbed dose of 6 kGy. Two confirmed experiments at optimal test conditions also performed 99.5% atrazine removal and were well fit with predicted results. Moreover, the operation cost at the optimal condition was determined approximately 3.032 \$/m^3, which was much cheaper than conventional treatment techniques. These obtained results highlight the potential of using the electron beam process to degrade atrazine contaminants.

Key words: Atrazine, herbicide, Electron beam, Taguchi design

INTRODUCTION

Atrazine belongs to the s-triazine derivative family of herbicides and is the most widely used pesticide to control pests and disease carriers[1]. Atrazine has strong aromaticity and high resistance to biological degradation[2]. Although atrazine is no longer used, it can still be found in various natural streams due to previous widespread use[3]. Phyu, Warne[4] found that atrazine was moderately toxic to tropical freshwater *daphnia* species (48-h-LC_{50} 24.6 mg/L). Atrazine can hydrolyze quickly in an acidic or basic environment, but it is relatively resistant to hydrolysis at neutral pH levels. In freshwater, atrazine is hydrolyzed with a half-life of 742 days and biodegraded after 40 days, while the half-life of atrazine in water is 60 days[5]. Conventional techniques such as adsorption, coagulation, filtration, or biological had been applied to militate atrazine from the environment[6–9]. However, due to their aromatic nature, these conventional methods are insufficient to eradicate herbicides from the environment. Processes based on hydroxyl radicals ($^{\bullet}OH$) with 2.80 V as the oxidation potential[10] are considered promising for the rapid degradation of pesticide pollutants. Fareed, Hussain[11] stated that Fenton reagent could remove 79.93% of atrazine in groundwater and with a couple of UV/ Fenton the removal could reach 97.02%. Only 42.57% of atrazine was removed in the case of UV. Even the combination of Fenton and UV could accelerate removal efficiency. However, the technique is constrained by energy and sludge treatment costs. Therefore, they could not be used for pesticide contaminant treatment. Among the AOPs, electron beam (EB) is considered a promising process to eliminate pesticides in the environment because it can rapidly degrade resistant organic compounds with less sludge production and no chemical requirement[12]. The decomposable capacity of EB in water could result from the formation of oxidizing species ($^{\bullet}OH$, H_2O_2, $HO_2^{\bullet}$…) and reducing species (e^-_{aq} and $H^{\bullet}$) through water radiolysis as following Equation 1[12].

$$H_2O \rightsquigarrow 2.8\,^{\cdot}OH + 2.7e^-_{aq} + 0.6H^{\cdot} + 0.72H_2O_2 + 2.7H_3O^+ + 0.48H_2 \quad (1)$$

EB has successfully removed a myriad of refractory organic pollutants from contaminated wastewater, such as pesticide[13], urban wastewater[14], textile wastewater (15), slaughterhouse wastewater[15,16]

Cite this article : Tan L V, Duy N N, Huong D T G, Ha B M. **Electron beam induced degradation of atrazine in solution using Taguchi approach**. *Sci. Tech. Dev. J. - Sci. Earth Environ.;* 5(2):417-423.

and pharmaceuticals[12,17]. Although a lot of EB has been investigated with hazardous organic compounds, scarce literature is still available to eliminate atrazine from aqueous solutions, especially treatment costs incurred using EB technique[12,13].

Aside from choosing removal tactics, experimental design is important for minimizing wastewater and wastewater treatment time and cost. The Taguchi method is one of the most uncomplicated cases of experimental design involving the minimum number of experiments to be performed within the permissible limit of factors and levels through the Signal to Noise ratio (S/N). The Taguchi design has wide application in multiple wastewaters, i.e., textile[18], pulp and paper mill[19], oily[20], etc. However, the application of the Taguchi method to the elimination of atrazine using the EB method is still scarce.

Hence, in the present study, the degradation of atrazine from aqueous solutions is studied using EB irradiation followed by the Taguchi approach. The variables are initial atrazine concentration, initial pH, H_2O_2 dosage and absorbed dose to obtain the best conditions for efficient atrazine degradation. The treatment cost was also evaluated to determine the potential of the EB process.

MATERIALS AND METHODS

Reagents

Atrazine (2-Chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine) with a purity of >98% was purchased from Sigma Aldrich, while other analytical grades of chemicals such as H_2SO_4, NaOH, H_2O_2, etc. were procured from Biochem (France). The 1000 mg/L of atrazine stock solution was prepared as our previous procedure[21] using deionized water and stored at 5°C. Freshly prepared distilled water was used for the preparation of the atrazine solution of the desired concentration from the stock solution.

Experimental setup and procedure

The EB irradiation procedure was performed according to our previous study[12]. Briefly, 1000 mL of specific atrazine solution at a desired pH value were put in a plastic box (solution thickness of 2.5 cm). A volume H_2O_2 then was added to the box to reach a the needed dosage; after that, the box was irradiated at specific absorbed doses (0.5 to 5.0 kGy, corresponding to 150 microseconds to 1.5 seconds) in an electron accelerator UERL–10–15S2 (10 MeV, 7.5 kW) at the Research and Development Center for Radiation Technology, Vietnam Atomic Energy Institute, Ho Chi Minh City. The experiments are based on the L9 Taguchi design with S/N to determine the optimal conditions for the atrazine removal via the EB process. Four independent variables were applied: initial pH (X_1), atrazine concentration (X_2), H_2O_2 dosage (X_3) and absorbed dose (X_4). The level of each code ranged from low (-1) to high (1) based on our previous studies on the degradation of atrazine[22] and EB[12], as shown in Table 1.

Analysis and statistical method

Dichromate dosimetry[23] was employed to measure absorbed doses during EB irradiation. While the atrazine concentration was determined using the High-Performance Liquid Chromatography (HPLC) Alliance 2695 model from Waters Corporation (Pennsylvania, USA) with the following parameters: wavelength of 224 nm, C18 column, length and diameter of column 4.6 × 250 mm, and injection volume of 20 μL. The percentage of atrazine removal was calculated as follows:

Atrazine removal efficiency = $\frac{C_0 - C_t}{C_0} \times 100\%$ (2)

Herein, C_0 is the initial atrazine concentration and C_t is the atrazine concentration at t reaction time.

The energy consumption (E_{con}) of EB is computed according to absorbed dose (D) and radiation energy utilization efficiency (f), which are typically 0.5 for UELR-10-15S2 electro accelerator as Equation 3 follows[13].

$$E_{con} = \frac{D}{3.6 \times f} \quad (3)$$

Treatment costs in this study only involve energy consumption and H_2O_2 costs. Given the Vietnamese market in July 2021, the electrical energy price is 0.065 \$/kWh and the H_2O_2 -50% price is 2.0 \$/ liter. Therefore, treatment costs are calculated by Equation 4.

Treatment cost (\$/m^3) = 0.0065 × E_{con} + 2 × $V_{H_2O_2}$ (4)

Where $V_{H_2O_2}$ could be determined by Equation 5.

$$V_{H_2O_2}\,(liter) = \frac{C_{M_{H_2O_2}} \times M_{H_2O_2}}{50 D_{H_2O_2}} \quad (5)$$

Here $C_{M_{H_2O_2}}$, $M_{H_2O_2}$ and $D_{H_2O_2}$ are optimal concentration (mM), molecular weight (34.0147 g/mol) and density (1.45 g/mL) of H_2O_2.

Taguchi results were evaluated by the "the-Larger-The-Better" as Equation 6.

$$S/N\,(dB) = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i}\right) \quad (6)$$

In which y_i is the results of each experiment and n: number of experiment .

The results were displayed as mean ± SD and the statistical software is Minitab 18.1 version (Minitab Inc, USA).

Table 1: Taguchi design for EB treatment

Independent factors	Level 1	Level 2	Level 3
Initial pH - X_1	5	7	9
Atrazine concentration - X_2 (mg/L)	2	4	6
H_2O_2 dosage -X_3 (mM)	1	3	5
Absorbed dose -X_4 (kGy)	2	4	6

RESULTS AND DISCUSSION

Experimental Design Analysis

According to the L9 Taguchi approach, nine experiment results with 3 levels and 4 factors are indicated in Table 2. The output signal–noise (S/N) ratio from the Taguchi analysis would be evaluated for each test run to determine the distinguishing characteristics between control and signal factors to optimize the pesticide removal procedure. The higher the S/N ratio, the more sufficient information there is compared to noisy erroneous data. The "larger, the better" of S/N was also used to evaluate the maximize pesticide removal efficiency of the EF process.

Minitab analysis of the Taguchi design

The influences of variables such as initial pH, H_2O_2 dosage, atrazine concentration, and absorbed dose to mitigate atrazine were studied. The efficiency of mitigation of atrazine was evaluated by the ranks of means and S/N ratios depicted in Figure 1 and Table 3.

As seen in Figure 1, the mean of the S/N ratios of each factor corresponds to their level. The higher the S/N ratio indicates the higher the results. Level 1 of pH[5] and atrazine concentrations (2 mg/L), level 2 of H_2O_2 dosage (3 mM) and level 3 of absorbed dose (6 kGy) display the best value for S/N ratio to mitigate atrazine using EB irradiation. The increased absorbed dose could increase the • OH formation and lead to an increase in the removal of atrazine[12]. While the higher atrazine contamination requires more oxidants for removal, high atrazine concentration negatively affected the treatment capacity[8,9]. Previous studies[12,15,16] demonstrated that EB capacity could be enhanced at the acid condition and add a small amount of H_2O_2 due to improving the number and the potential oxidant capacity of $^{\bullet}OH$ by Equation 7. However, exceeding H_2O_2 dosage could reduce • OH concentration and decrease the atrazine removal capacity (Equation 8).

$e^{-}_{aq} + H_2O_2 \rightarrow {}^{\cdot}OH + OH^{-}$ (7)

$H_2O_2 + {}^{\cdot}OH \rightarrow 2H_2O + O_2$ (8)

These results are consistent with the previous study; they stated that most organic compounds were predominately degraded as coupled with H_2O_2 and small pollutant concentrations with specific absorbed values at acidic conditions[13,14,24].

Based on the S/N ratio in table 3, the "Predict Taguchi result" had been conducted to find the best conditions for atrazine removal at an initial pH of 5, an atrazine concentration of 2 mg/L and an H_2O_2 dosage of 3 mM at an absorbed dose of 6 kGy. The predicted result shows that most atrazine is eradicated at these combination conditions (100.1%) with S/N of 40.1022. Two verify experiments at these conditions had demonstrated the efficiency of EB treatment with a removal efficiency of 99.5%. These results again proved the fitness of the predicted model of Taguchi design.

Cost analysis

The best conditions for removing most of the atrazine using EB are at an initial pH of 5 (the natural pH of atrazine solutions), an atrazine concentration of 2 mg/L and an H_2O_2 dosage of 3 mM at an absorbed dose of 6 kGy. The energy consumption is computed by Equation 2 and gave the result of 3.33 kWh while the volume of H_2O_2 is 1.407 L calculated using Equation 4. The treatment cost is computed at approximately 3.032 $\$/m^3$ (Equation 3). This cost was much cheaper than 20.91 $\$/m^3$ in Gaied, Louhichi[25]; they were using EF for treating domestic wastewater or 10.68 $\$/m^3$ for landfill leachate samples[26].

CONCLUSION

Most of the atrazine from the aqueous solution (99.5%) was eliminated using EB irradiation with the Taguchi approach. The best removal efficiency was reached at pH 5, atrazine concentration of 2 mg/L, H_2O_2 dosage of 3 mM and absorbed dose of 6 kGy. The theoretical prediction optimizer tool in Minitab released a treatment efficiency of 100.1%, consistent with the obtained results from two verified experiments (99.5%). At optimal condition, the EB treatment cost was approximately 3.032 $\$/m^3$, cheaper

Table 2: Experimental design, the obtained responses

Run	Initial pH	Atrazine concentration (mg/L)	H_2O_2 dosage (mM)	Absorbed dose (kGy)	Atrazine removal (%)	S/N
1	5	2	1	2	83.2	38.4025
2	5	4	3	4	95.8	39.6273
3	5	6	5	6	90.4	39.1234
4	7	2	3	6	98.5	39.8687
5	7	4	5	2	81.6	38.2338
6	7	6	1	4	82.7	38.3501
7	9	2	5	4	89.3	39.0170
8	9	4	1	6	91.8	39.2569
9	9	6	3	2	75.2	37.5244

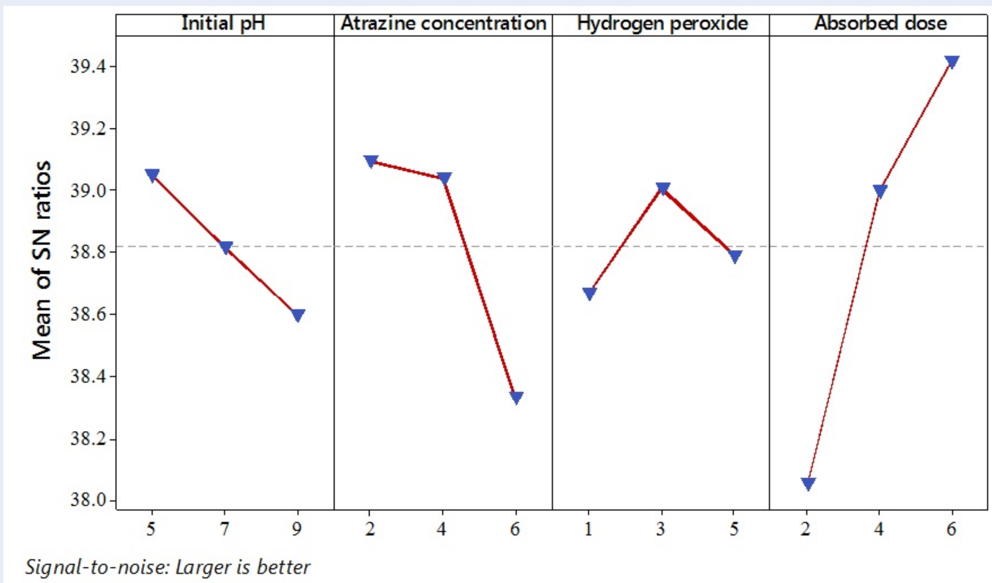

Figure 1: The effect of variables on the S/N for mitigating atrazine

than other AOPs treatments. This study demonstrated EB could be an exemplary process for pesticide contamination treatment.

ACKNOWLEDGMENTS

This work was supported by the National Foundation for Science and Technology Development (NAFOSTED) under Project No. 105.08-2019.17.

ABBREVIATION

AOPs: Advanced Oxidation Processes
COD: Chemical Oxygen Demand
EB: Electron beam
S/N: Signal/Noise

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this article

AUTHORS' CONTRIBUTIONS

Luu Van Tan, Nguyen Ngoc Duy, Duong Thi Giang Huong and Bui Manh Ha have made substantial contributions to the work reported in the manuscript.

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Table 3: S/N ratio and mean response

Items	Level	Initial pH	Atrazine concentration (mg/L)	H_2O_2 dosage (mM)	Absorbed dose (kGy)
S/N ratio	1	39.05	39.10	38.67	38.05
	2	38.82	39.04	39.01	39.00
	3	38.60	38.33	38.79	39.42
	Delta	0.45	0.76	0.34	1.36
Mean	1	89.80	90.33	85.90	80.00
	2	87.60	89.73	89.83	89.27
	3	85.43	82.77	87.10	93.57
	Delta	4.37	7.57	3.93	13.57
Rank	Rank	3	2	4	1

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Tạp chí Phát triển Khoa học và Công nghệ – Khoa học Trái đất và Môi trường, 5(2):417-423

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Bài nghiên cứu

Giảm atrazine trong nước bằng phương pháp chùm tia điện tử sử dụng thiết kế Taguchi

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TÓM TẮT

Nghiên cứu này xử lý atrazine trong nước giả lập bằng phương pháp chiếu xạ chùm tia điện tử một phương pháp xử lý bậc cao được sử dụng trong xử lý môi trường thời gian gần đây. Atrazine với nồng độ từ 2 đến 6 mg/L pha trong nước được chọn làm đối tượng xử lý khi thay đổi chùm tia điện (2- 6 kGy) tại pH thay đổi từ 5 đến 9. Nghiên cứu này cũng kết hợp chùm tia điện tử với H_2O_2, khi được thêm vào trong quá trình xử lý từ 1 đến 5 mM. Thí nghiệm xử lý tuân theo thiết kế Taguchi một thiết kế đơn giản được đánh giá thông qua tỷ lệ tín hiệu trên độ ồn. Kết quả thí nghiệm cho thấy liều chiếu xạ, hàm lượng atrazine và pH là các yếu tố ảnh hưởng lớn đến hiệu quả loại trừ atrazine, trong khi hàm lượng H_2O_2 ít ảnh hưởng đến khả năng loại trừ atrazine. Tính toán lý thuyết cho thấy hầu hết atrazine (100.1%) bị loại trừ tại liều chiếu xạ 6 kGy, nồng độ atrazine là 2 mg/L, hàm lượng H_2O_2 3 mM tại pH 5. Thí nghiệm kiểm chứng tại điểm thí nghiệm tối ưu này cho thấy 99.5% atrazine bị loại trừ, tại điểm tối ưu này chi phí xử lý atrazine bằng phương pháp chiếu xạ khoảng 3,032 USD/m^3. Kết quả này cho thấy tiềm năng của việc loại trừ các hợp chất ô nhiễm hữu cơ bền như thuốc bảo vệ thực vật bằng phương pháp chùm tia điện tử.

Từ khoá: Atrazine, thuốc trừ cỏ, Chùm tia điện tử, thiết kế Taguchi

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Lịch sử

- Ngày nhận: 17-8-2021
- Ngày chấp nhận: 17-9-2021
- Ngày đăng: 07-11-2021

DOI : 10.32508/stdjsee.v5i2.653

Trích dẫn bài báo này: Tấn L V, Duy N N, Hương D T G, Hà B M. **Giảm atrazine trong nước bằng phương pháp chùm tia điện tử sử dụng thiết kế Taguchi**. *Sci. Tech. Dev. J. - Sci. Earth Environ.;* 5(2):417-423.