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Research Article

Optimization of Atrazine Degradation in the Aqueous Phase Using Titanium Catalyst Doped With Iron (Fe^{+3}-TiO_2) Processes

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Abstract

Background: Atrazine is used extensively in many parts of the world for controlling a variety of weeds, primarily in the production of corn. Its widespread use in water has caused environmental concern because of its frequent detection in aquatic systems where this herbicide has been spilled.
Objectives: The purpose of this study was to focus on evaluating the feasibility of using titanium catalyst doped with iron (Fe^{+3}-TiO_2) to remove atrazine from the aqueous phase and to determine its removal efficiency in ideal conditions.
Materials and Methods: The influence of different parameters was considered. Three different levels of pH (3 - 11), initial concentration of atrazine at three different levels (0.1 - 10 mg/L), and reaction time at five different levels (0 - 120 minutes) in the removal of atrazine in an aqueous-phase titanium catalyst doped with iron (Fe^{+3}-TiO_2) at three different levels (0 - 25) were investigated. Finally, the data were analyzed using SPSS software (version 16).
Results: The results demonstrated that the atrazine removal rate increased by decreasing the initial atrazine concentration and the maximum rate of atrazine removal (41.72%) was at pH = 11, atrazine concentration = 0.1 mg/L, and catalyst concentration = 25 at 120 minutes.
Conclusions: According to the results, it can be concluded that the titanium catalyst doped with iron (Fe^{+3}-TiO_2) processes is an appropriate method for reducing atrazine in polluted water resources.

Keywords: Atrazine, Liquid Solution, Catalyst, Removal

1. Background

Atrazine (2-chloro-4-ethylamino-6-isopropylamino-1, 3, 5-triazine) is a herbicide used for weeds pre- and post-emergence control, particularly in maize crops (1). It is primarily used to kill weeds in agriculture, but it has also been used for highways, railroads, forestry, orchards, industrial weed control, private use in gardens, and as a biocide (algal control). Due to its low cost, atrazine has rapidly become one of the most common herbicides in agriculture, especially in maize crops (2).

Among pesticides, atrazine is widely used and is included on the European commission's priority substances list (3, 4). environmental protection agencies (EPA)have estimated that between 34 and 36 million kilograms of atrazine were used in 2001 (5). Thus, it is frequently detected in underground water sources, drinking water supplies, and runoffs (4, 6), and it seriously influences water quality (7). Due to its bio-resistance, atrazine remains in the soil and aquatic environment (1); this causes serious environmental problems. Moreover, it penetrates through surface and subsurface water bodies due to its excessive usage and high persistence and mobility (8, 9).

Atrazine is slightly toxic for humans, animals, and plants, since it can be absorbed by inhalation, ingestion, or through the skin (10). Therefore, environmental concentrations of atrazine should not exceed 20 g/L, except in small temporary puddles in fields, where peak concentrations can be greater than 200 g/L for short periods of time after a flooding (11).

The international agency for research on cancer (IARC) has concluded that atrazine is a type 2B carcinogen. The maximum contaminant level (MCL) for atrazine in drinking water established by the (EPA) is at 3.0 $\mu g.L^{-1}$ (12). When people are exposed to a high level of atrazine above the MCL in a relatively short period of time, they may face congestion of the heart, lungs, and kidneys; low blood pressure; muscle spasms; weight loss; and damage to the adrenal gland (8).

In recent years, more limits have been imposed in the use of pesticides and herbicides like atrazine in agriculture; they have been recognized as potential and very persistent pollutants for aquatic systems (13). Still, advanced methods are needed for the effective treatment of pesticide-polluted ground and surface water (14). Various techniques have been suggested for the elimination of atrazine and related compounds from drinkable water, including biological degradation (8, 15, 16), adsorption (15, 17), oxidation by ozone, or the Fenton reaction, direct and catalyzed photolysis, sonolysis, and radiolytic decomposition. Amongst the photocatalysts tested, titanium dioxide is particularly efficient (18).

In recent decades, TiO_2 has been widely used as a photocatalyst for the treatment of pollutants due to its high photocatalytic activity, high chemical stability, low toxicity, and so on. It can disintegrate pollutants completely to carbon dioxide and inorganic constituents in both the solution and gas phases. However, TiO_2 is not active enough in photocatalysis in the range of visible irradiation. Over the last decade, the improvement of the TiO_2 catalyst has received a great deal of attention. Several research attempts have involved doping it with a small amount of expensive metals and metal ions, after which it exhibited higher absorbance in visible irradiation (18, 19).

Some researchers have studied the photocatalytic activity of TiO_2 in the visible light region. It can only make use of 3% - 5% of the solar spectrum that reaches the earth because of its large band gap (Ebg, anatase $\cong$ 3.2 ev, rutile $\cong$ 3.0 ev) (7); thus, it is essential to produce visible light-responsive (VLR) TiO_2, not only to use visible light but also to reduce the recombination of photogenerated electrons and holes (19, 20).

A study by Bianchi et al. showed that in the presence of 0.1 g of TiO_2 and irradiation wavelengths of 315 - 400 nm, 37% atrazine degradation occurred (13). Safari and his coworkers showed that the photocatalytic activity of iron-doped TiO_2 was greater than TiO2, because of the smaller crystal size, and higher light absorption ability (19). Moreover, Mirovic et al. showed that during 2 hours of irradiation of atrazine solution (1 mg/L) with 5 g TiO_2 (0.05%) alginate (1.5%) micro-beads, it was observed that by the end of the experiment, atrazine was being degraded up to 98%, and that the highest level of degradation was attained after 45 minutes (21).

Atrazine is characterized by its persistence remained in environment for days and years. Its persistence is due to the stability of the s-triazine ring It has been reported that atrazine has endocrine-disrupting capabilities. Thus, the herbicide can have dangerous side effects in the environment and for humans, and we need to apply different methods to control its usage and find effective solutions to eliminate its residues from underground water sources. Many studies have been conducted on the removal of atrazine using the Advanced oxidation process AOP process. However, to date, no studies applied fortified titanium catalyst doped with iron (Fe^{+3}-TiO_2) for the removal of atrazine.

Here, the sol-gel method was used to prepare Fe^{+3}-TiO_2 nanocatalyst powder. At first, ferrous nitrate was dissolved in half of propanol (121.775 mL) and completely mixed. After 15 minutes, 121.775 mL of propanol was mixed with 62.77 mL of titanium tetraisopropoxide (TTIP), and then the mixture was added very slowly to the former solution over 75 minutes to form the sol. Meanwhile, deionized distilled water (8.33 mL) was added to the solution as well. Thirty minutes after the addition of propanol to TTIP, the pH was adjusted to 3 by nitric acid. All the processes were carried out in mixing mode using a homogenizer. Then, the resulting solution was placed on the magnetic mixer for 24 hours to form jelly. This was put in the oven at 80°C for 10 hours to evaporate the alcohol. To activate the catalyst, the jelly was placed in oven at 500 ± 50°C for 2 hours. The activated catalyst was put in a desiccator until it was cool. Finally, the catalyst was powdered (22).

2. Objectives

In the present study, TiO_2 with iron (Fe^{+3}) was selected as a catalyst in the photocatalytic removal of atrazine. The effects of several operational parameters on the photocatalytic degradation of atrazine, including pH, Fe^{+3}-TiO_2 dosage, and atrazine concentration, were investigated.

3. Materials and Methods

The studied parameters were reaction time (0, 30, 60, 90, and 120 minutes), TiO_2 with iron (Fe^{+3}) concentration (0 - 25 mg/L), and initial atrazine concentration (0.1, 1, and 10 mg/L) at different pH levels (3 - 11). The optimal conditions were determined according to the maximum atrazine removal efficiency (23). The data were analyzed using SPSS for windows (version 16, SPSS Inc., Chicago, IL, USA). One factor at the time of the designing was used for analysis of the parameters and the interaction of the factors was studied. All of the data were presented based on the mean.

3.1. Chemicals and Analytical Method

Atrazine with 99.9% purity was purchased from Sigma Aldrich company (USA). Other chemical products such as TiO_2 were purchased from Merck (Germany). We used a scanning electroscope microscope (SEM; EM3200, KYKY company, China) to determine the morphology and mean diameter of the catalyst's particles. Fortified nanoparticles of titanium dioxide (TiO_2) were doped with trivalent iron according to the sol-gel method (22).

For atrazine detection in the aqueous phase, a waters model high performance liquid chromatography (HPLC; Waters YL9100HPLC SYSTEM, USA) system with C18 columns (CP-SIL 5 CB column model, 250 × 4.6 mm, 5 μm) was calibrated and tested prior to the injection of samples. The mobile phase included methanol and water (20/80 V/V), with a flow rate of 0.5 mL/minute. An ultraviolet (UV) absorbance detector at a wavelength of 224 nm was used to detect atrazine in the samples. The retention time for atrazine was 9 minutes. The detection limit for the sample was 0.001 mg/L. The atrazine chromatogram is presented in Figure 1.

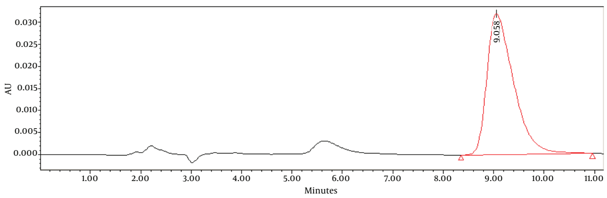


Figure 1. Atrazine Chromatogram

3.2. Reactor Specifications

The specifications of the photochemical reactor are shown in Figure 2. The experiment was performed in a 1 L volume reactor. The test was performed in a closed glass reactor with an adjustable mixer.

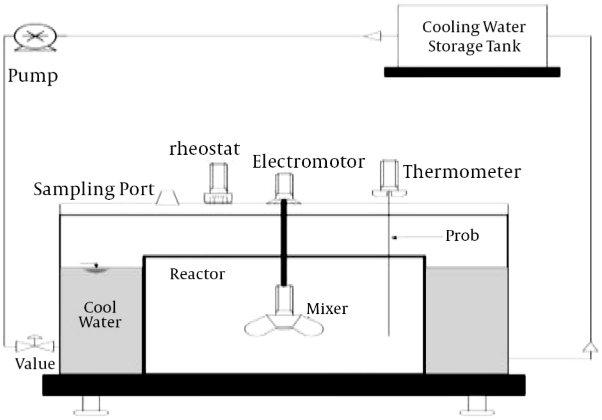


Figure 2. The Specifications of the Photochemical Reactor

The atrazine removal efficiency was calculated using the following formula:

$$E\,(\%) = \left(\frac{A_0 - A}{A_0}\right) \times 100 \qquad (1)$$

Where A_0 and A represent the atrazine concentration before and after the titanium catalyst doped with iron (Fe^{+3}-TiO_2) process and E represents the efficiency.

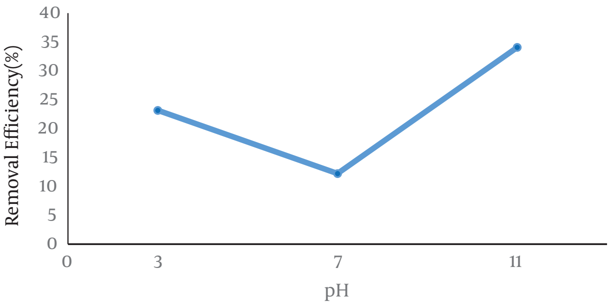
3.3. Effects of pH, Catalyst Concentration, Initial Atrazine Concentration, and Reaction Time on the Removal Rate of Atrazine by the Titanium Catalyst Doped With Iron (Fe+3-TiO2)

To measure the influence of different parameters on the removal rate of atrazine by the titanium catalyst doped with iron (Fe^{+3}-TiO_2) in the aqueous phase, different parameters were studied, such as the pH, initial atrazine concentration, catalyst concentration, and reaction time. Ammonium hydroxide and hydrochloric acid were used to adjust pH in the samples (24). The samples were passed through a Whatman filter cellulose acetate membrane with a pore size of 0.22 microns (Germany). Following this, the atrazine residual was determined by HPLC. All of the experiments were performed two replications and in the presence of the control samples.

4. Results

Photodegradation of atrazine using the titanium catalyst doped with iron (Fe^{+3}-TiO_2) is shown in Figures 3 - 6.

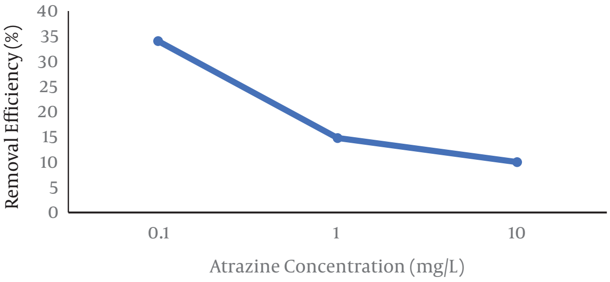
Figure 3. Effect of pH on the Atrazine Removal Efficiency Rate Using the Fe^{+3}-TiO_2 Process



Initial atrazine concentration = 0.1 mg/L, Fe^{+3}-TiO_2 = 25 mg/L, reaction time = 30 minutes.

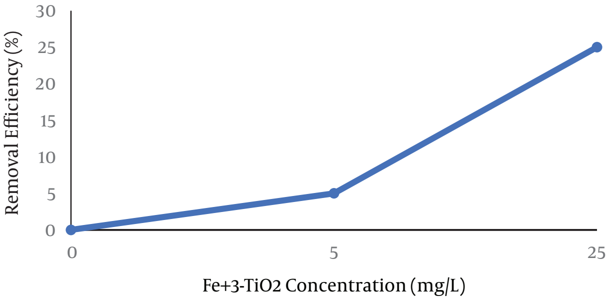
According to Figure 3, as the pH increased from 3 to 7, the atrazine removal rate decreased with a slightly milder angle. Then, when the pH increased from 7 to 11, the removal rate increased at a sharper angle. The result of this research showed that the maximum removal rate of

Figure 4. Effect of the Initial Atrazine Concentration on the Atrazine Removal Efficiency Rate Using the Fe^{+3}-TiO_2 Process



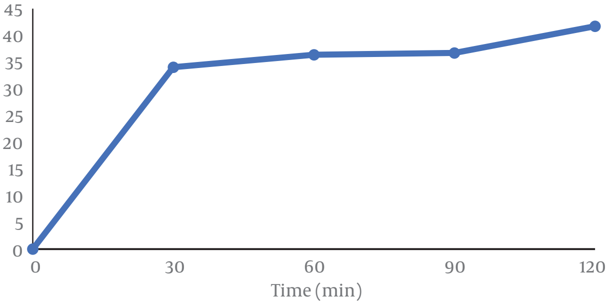
pH = 11, Fe^{+3}-TiO_2 = 25 mg/L, reaction time = 30 minutes.

Figure 5. Effect of Fe^{+3}-TiO_2 Concentration on the Atrazine Removal Efficiency Rate Using the Fe^{+3}-TiO_2 Process



pH = 11, initial atrazine concentration = 0.1 mg/L, reaction time = 30 minutes.

Figure 6. Effect of Reaction Time on the Atrazine Removal Efficiency Using the Fe^{+3}-TiO_2 Process



pH = 11, initial atrazine concentration = 0.1 mg/L, Fe^{+3}-TiO_2 = 25 mg/L.

atrazine (34.05%) occurred in an alkaline condition (pH = 11).

As shown in Figure 4, as the initial atrazine concentration increased from 0.1 to 1 mg/L, the atrazine removal rate decreased at a sharper angle. Then, by increasing the initial atrazine concentration from 1 to 10 mg/L, the removal rate decreased again but at a slightly milder angle. The results of this research showed that as the Fe^{+3}-TiO_2 concentration increased from 5 to 25 mg/L, the removal efficiency of atrazine increased.

Based on Figure 6, increasing the reaction time resulted in an increase in the removal efficiency. The removal efficiency increased from 34.05% after 30 min of reaction time to 41.72% after 120 minutes.

5. Discussion

In the present research, the optimization of the influential parameters, including the pH, initial atrazine concentration, catalyst concentration, and reaction time on removal of atrazine herbicide from the aqueous phase using titanium catalyst doped with iron (Fe^{+3}-TiO_2) were studied.

5.1. Effect of pH on Atrazine Removal

pH is one of the most important factors influencing the efficiency of many chemical and biological processes (12, 25, 26). Our data showed that the maximum removal rate of atrazine (41.72%) occurred at pH = 11 due to the formation of high concentrations of hydroxyl radical (27). Hemmati et al. used a Fe^{+3}-TiO_2/ UV-A process in the aquatic phase in the photocatalyst degradation of phenol and showed that the efficiency rate is best at pH = 3 (62.4%). This indicates the important role of pH in the UV/photocatalyst process (28).

Dehghani et al. demonstrated that there was a significant difference between pH and atrazine biodegradation (12), and many other studies have shown that better removal of atrazine occurs at higher pH levels (10, 29). Using an ozonation process, Prado et al. also showed that the maximum removal rate of atrazine is at pH = 11.5, and the removal rate of atrazine increased as pH increased (29). The result of this research agrees with that of the studies performed by Mamian et al. (10) and Prado et al. (29).

5.2. Effect of the Initial Atrazine Concentration on the Atrazine Removal Efficiency

The initial concentrations used in this research were 0.1, 1.0, and 10 mg/L. As shown in Figure 4, the removal rates of herbicide at the initial concentration of 0.1, 1.0 and 10 mg/L were 34.05, 14.78, and 10.01%, respectively. According to the results, as the initial atrazine concentration increased from 0.1 to 1 mg/L, the atrazine removal rate decreased dramatically. Afterwards, by increasing the initial atrazine concentration from 1 to 10 mg/L, the removal rate decreased again but not significantly. Based on the data we obtained in the present study, an initial atrazine concentration of 0.1 mg/L is optimal for atrazine degradation (41.72%), because samples with lower initial atrazine concentration have a higher chance of removal (24). Many other studies showed that better removal of atrazine occurred at lower concentrations (30, 31). Dehghani et al. showed that maximum removal efficiency (74.98%) was observed with the minimum initial concentration of the dye (50 mg/L) (30). Hoseini et al. demonstrated that decreasing the initial concentration of trichloroethylene (TCE) caused an increase in the removal rate (31), which is in line with the results of this study. However, Baghapour et al. reported that by increasing the initial concentration of atrazine, the removal efficiency increased (8).

5.3. Effect of Fe^{+3}-TiO_2 Concentration on the Atrazine Removal: REACTION Time

In recent decades, the unique properties of titanium dioxide (TiO_2), such as its high photocatalytic activity, high chemical stability, and low toxicity, have made it a suitable photocatalyst (19). Figure 5 shows the atrazine removal efficiency with the mentioned parameters (pH = 11, initial atrazine concentration = 0.1 mg/L, reaction time = 30 minutes). The titanium catalyst doped with iron (Fe^{+3}-TiO_2) was more efficient in the removal of atrazine at Fe^{+3}-TiO_2 concentration of 25 mg/L. Our data showed that as the Fe^{+3}-TiO_2 concentration increased from 5 to 25 mg/L, the removal efficiency of the atrazine increased from 10% to 36%. This result is similar to those reported by other researchers (24, 28, 31). Hemati et al. used a UV/TiO_2-Fe^{+3} process in the aquatic phase in the photocatalyst degradation of phenol, showing that by increasing the concentration of the catalyst from 0.25 to 0.5 gr/L, the number of absorbed photons increased; this led to an increase in the number of activated sites accessible in photocatalyst surface, and the amount of absorbed phenol increased (28).

Hoseini et al. demonstrated that the TCE removal increased with an increase in TiO_2 concentration and reached a plateau at a TiO_2 concentration of 100 mg/L, after which it remained approximately stable (31). Dehghani et al. showed that the penicillin G photocatalytic removal rate increased as the applied catalyst (Fe^{+3}-TiO_2) dose increased from 30 to 90 mg/L (24).

5.4. Effect of Reaction Time on Atrazine Removal

The effect of the reaction time on the atrazine removal efficiency is depicted in Figure 6. Initially, the degradation of atrazine was rapid (30 minutes reaction times); then, it became slower until it reached a plateau of 120 minutes. Based on the results, increasing the reaction time resulted in an increase in the removal efficiency.

Dehghani et al. demonstrated that this phenomenon may be related to atrazine photo degradation when its exposure time to the UV-A radiation is increased, and that a greater oxidation reaction occurs in the presence of hydroxyl radicals (32). In fact, the reaction rate at equilibrium reached a plateau. Moreover, determining the required time to reach a balance is also very important (33, 34, 35, 36). Vlaardingerboek showed that atrazine was completely degraded using UV/TiO_2 after a reaction time of 150 minutes (2). Bahena et al. demonstrated that complete oxidation of atrazine occurred after 75 minutes by TiO_2 in the presence of sunlight (37).

5.5. Conclusion

This study showed that the titanium catalyst doped with iron (Fe^{+3}-TiO_2) can significantly reduce the concentration of atrazine in the aqueous phase. The results showed that different parameters (pH, initial concentration of atrazine, catalyst concentration, and reaction time) affected the removal rate of atrazine. The maximum removal rate (41.72%) was achieved at pH = 11, initial atrazine concentration = 0.1 mg/L, catalyst concentration = 25 mg/L, and reaction time = 120 minutes.

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Footnotes

Authors' Contribution: Conceived and designed the experiments, Mansooreh Dehghani, Mohammad Ali Baghapour, Simin Nasseri; contributed reagents/materials/analyses, Narges Shamsedini, Mansooreh Dehghani; wrote the manuscript, Mansooreh Dehghani, Narges Shamsedini, Mohammad Ali Baghapour, Simin Nasseri, Mohammad Sohrab Moghaddam.

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