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Removal of Paracetamol from Pharmaceutical Waste Water by using UV/TiO2 and UV/ZnO2

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ABSTRACT

Pharmaceutical waste is one of the major complex and toxic industrial wastes. Pharmaceutical industry produces a wide variety of products. This industry uses both inorganic and organic as raw materials the latter being either of synthetic or of vegetable and animal origin. Antibiotics and vitamins are produced by fermentation of fairly complex nutrient solutions of organic matter and inorganic salts by fungi or bacteria. Photocatalytic degradation might be facilitated by using semiconductor ZnO and TiO_2 which is act to increase the surface area of paracetamol solution to exposed UV light which resulted increase photocatalytic degradation efficiency.

The effect of contact time on % reduction of paracetamol using UV/TiO_2 process. The experimental analysis carried out for 5 ppm and 10 ppm solutions. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 40, 57, 68, 73 and 80 resp. using UV/TiO_2 process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 29, 50, 64,69 and 72.5 resp. using UV/TiO_2 process. As per analysis its clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

The effect of contact time on % reduction of paracetamol using UV/ZnO process. The experimental analysis carried out for 5 ppm and 10 ppm solutions. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 30, 50, 62.5, 71.5 and 77 resp. using UV/ZnO process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 25, 42, 57.5 and 69 resp. using UV/ZnO process. As per analysis it's clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

Introduction

Due to rapid growth of population and industrialization the requirement of water increa se but the natural source of water which is useful for the domestic and industrial uses is very limited. From the industrial process the large amount of waste water is coming out treatment of this waste is necessary to protection of environment and human being from harmful effect.

Availability of water is becoming an increasing concern in the globalized world, both in developed and in developing countries. A sustainable use of water sources could result in the search of additional water sources or even in recycling wastewater treatment plant effluents. The goal of biological wastewater treatment is a stepwise oxidation of organic pollutants aiming to achieve complete mineralization.

Numerous wastewater constituents are persistent to biodegradation or they are only subjected to minor structural changes instead of complete transformation into carbon dioxide and water. They may be eliminated by applying advanced biotic treatment processes such as membrane filtration, UV degradation, ozonation, advanced oxidation processes, one of them being cavitation.

Pharmaceutical waste is one of the major complex and toxic industrial wastes. Pharmaceutical industry produces a wide variety of products. This industry uses both inorganic and organic as raw materials the latter being either of synthetic or of vegetable and animal origin. Antibiotics and vitamins are produced by fermentation of fairly complex nutrient solutions of organic matter and inorganic salts by fungi or bacteria.

If a crude waste from an antibiotic plant is discharged into a stream it not only imparts objectionable odor to the stream but also adversely affects the flora and fauna. The volume and composition of the liquid waste not only vary from plant to plant but also from section to section in a plant, producing different type of drugs from raw materials and using varieties of processes.

LITERATURE REVIEWS

Photocatalytic degradation might be facilitated by using semiconductor ZnO which is act to increase the surface area of paracetamol solution to exposed UV light, which might be resulted in reaching to photocatalytic degradation efficiency (P.D.E) equal to 100 % at 30 min for paracetamol. Both UV light and catalyst are important factors in paracetamol degradation. the initial concentration increase for both drugs leads to reduce degradation rate with foundation of the mentioned weights of ZnO. [1].

The increase of pH of reaction solution for both drugs have a positive effect by increasing the degradation rate until the pH 8 and 4 are the best acidic function of the paracetamol and procaine. Zinc oxide with 99% purity and solutions were prepared using double distilled water. The required amount of the catalyst suspended in 200 ml of aqueous solutions of drugs using a magnetic stirrer. The pH of the solution (pH 2–12) and concentration of drugs (5, 10, 20, 30 and 40) g/l. the reaction temperatures amounted to 15, 25, 35, 45, and 55 ^{O}C. [1].

Optimal temperatures is 55 ^{O}C for paracetamol and procaine with initial concentration 10 gm/l for both paracetamol and procaine, ZnO amount 0.025 gm for paracetamol and 0.1 gm for procaine, and pH solution 8 and 4 for paracetamol and procaine respectively. The effect of pH on photocatalytic degradation rate and optimal pH is 8 for paracetamol and 4 for procaine in presence of certain conditions [1].

Tetracycline (TCs) in waste water utilizing Zinc oxide (ZnO) Nano particle as catalyst. The effects of several factor like influence of primary Tetracycline (TCs) drug concentration, irradiation time and the effect of catalyst ZnO. Tetracycline (TCs) drug at 0.2 gm / 100 cm^3 weight of (ZnO) Zinc oxide Nano particle and 50 mg /l of Tetracycline (TCs) drug. The percentage capacity of drug removal at 80.388 %. The substance about 0.5 – 3 gm/l in the solution by 50 mg/l concentration drug at time 1 hr. at temp 25 ^{O}C. 100 ml of pharmaceutical contaminants and add 2 g/L of TiO_2 nanoparticle in beakers and put under UV. [2].

The Photocatalytic degradation of the Esomeprazole and Azithromycin antibiotics in aqueous solution under UV irradiation and TiO_2 nano powder. Percentage Degradation of this antibiotics improved when Titanium dioxide concentration increased. The degradation found maximum when TiO_2 0.5 -1.0 gm/L. At a room temperature and pH 6 the 60 % COD reduction achieved with a 150 min. 67% COD

reduction achieved in a case of Azithromycin. COD removal increased with increasing TiO_2 concentration and decreasing initial concentration of antibiotics. [3]

Photo-catalysis enhanced by UV irradiation appears to be a promising technology for the degradation of pharmaceuticals in wastewater. TiO_2 catalyst used and prepared by sol-gel techniques .Photocatalytic treatment applied to the degradation of Azithromycin and Esomeprazole antibiotics in the aqueous solution. Amount of catalyst is important for the degradation of antibiotics. [3]

AOP like Photocatalytic technique use for the removal of organic pollutants from pharmaceutical wastewater streams by using Titanium dioxide. Photocatalytic degradation reduces Chemical Oxygen Demand (COD) from wastewater. UV light was utilized in the experimental setup. UV light passes through mixture of TiO_2 powder and wastewater sample agitated with continuous stirring at room temperature. The photocatalytic removal of organics and its degradation efficiency evaluated by determination of reduction in the COD values. [4]

The effects of initial effluent concentration, catalyst dosage, irradiation time, effect of temperature and effect of pH were studied. Effluent and TiO_2 mixed continuously in a flask followed by stirring for 1h and the concentration of wastewater sample maintained at 10:6 ratio, optimum values of the variables the irradiation time 60 min, catalyst dosage 0.8 g/50 ml, temperature 35 ^{O}C and pH 12. The photocatalytic treatment to destruct organic pollutants in 1h and induced a (COD) reduction of 93.06%. UV light by varying the amount of Photocatalyst load from 10 mg to 100 mg and the illumination time fixed for 1h. When the amount of Photocatalyst load increased in the wastewater photocatalytic degradation efficiency significantly increased and degradation rate constant found at 80 mg of Photocatalyst load and removal efficiency 90.08 %. [4].

TiO_2 based nanocomposite photocatalysis process has presented effective degradation towards PhACs. Operational parameters such as dopant content, catalyst loading and initial pH were the major factors in the photocatalysis system. TiO_2 based nanocomposite for the degradation of PhACs. Advanced oxidation processes (AOPs) are considered the most effective processes as they present a significant potential for the treatment of a wide variety of emerging contaminants. [5]

Use of TiO_2 photocatalysis 82% of paracetamol removal obtained under optimum conditions and under 6 h of natural sunlight irradiation and 75% of tetracycline removal obtained under optimum conditions and under 30 min of natural sunlight irradiation. TiO_2 photocatalysis process for the removal of

pharmaceuticals focused on optimizing the operating parameters that can yield the highest degradation efficiency towards pharmaceuticals. [5]

The degradation of pharmaceuticals is highly dependent on the operational parameters of the system. Operating parameters such as initial pH, initial concentration of the pollutant, catalyst loading and irradiation time can influence the removal rate of pollutants. The photocatalytic degradation able to removal of pharmaceutical compounds such as ketoprofen, ibuprofen, tetracycline, amoxicillin and naproxen. [5]

The photocatalytic degradation of real pharmaceutical wastewater using TiO_2, ZnO and H_2O_2. The pretreated sample wastewater used for degradation at 38 ^{O}C under pH of 9 and 4 in a stirrer bath reactor equipped with eight ultraviolet tubes. Removal of organic, inorganic pollutants and pathogens. AOPs for removal of residual pharmaceuticals from real pharmaceutical wastewater. [7]

The three catalysts (Titanium dioxide, zinc oxide and TiO_2/ H_2O_2) used are effective catalysts in photocatalytic degradation of real pharmaceutical wastewater. The maximum degradation achieved 45.11% by combined use of TiO_2 and H_2O_2 at 38 ^{O}C and pH. The degradation improved at higher pH with Zinc oxide and Titanium oxide. Effect of these three catalyst can be testes at various pH and different contact time by experimental analysis.[7].

Pharmaceutical WWT with initial concentrations Ci > 0.3 mg/l present in a WWTP effluent. UVA (1.5 mW/ cm^2 and intensity peak at 365 nm) irradiation of TiO_2 P-25 (As = 56 m^2 g/l) or ZnO (As = 5.23 m2 g/l) nanoparticles leads to considerable degradation of pharmaceuticals. ZnO nanoparticles 40 min UVA irradiation sufficient to degrade over 95% of these pharmaceuticals. Using TiO_2 P-25. In this system ZnO presents faster degradation. Degradation of pharmaceuticals present in a real wastewater sample by photocatalysis conduct successfully via photocatalysis by ZnO and TiO2 under UVA-radiation. ZnO shows higher degradation rates and prevent ZnO contamination of the aquatic environment due to its potential toxicity. [8]

Color and COD degradation of 150-300 ppm of Procion Red by using 0.5-8 g/l of TiO_2 catalyst under solar irradiation. The higher of TiO_2 catalyst amount and the longer the exposure time use and higher color and COD degradation percentage. When using Procion Red of 150 ppm highest color degradation of 100% achieved by using TiO_2 catalyst of 6 g/l and the highest COD degradation of 62% obtained by using TiO_2 catalyst of 8 g/l under 12 hours of solar irradiation. [9].

Advanced Oxidation Processes (AOPs) may become the most widely used water treatment technologies for organic pollutants not treatable by conventional techniques due to their high chemical stability or low biodegradability. Processes involve generation and subsequent reaction of hydroxyl radicals (•OH) which are one of the most powerful oxidizing species. Oxidation processes like TiO_2/U, H_2O_2/UV, Photo-Fenton and ozone (O_3, O_3/UV, O_3/H_2O_2). AOPs which can be driven by solar irradiation, photo-Fenton and heterogeneous catalysis with UV/TiO^2. Photo-Fenton treatment mineralizes 80% of the TOC in the rinse water in a batch process. Advanced oxidation process (AOP) and is suitable for the oxidation of a wide range of organic compounds. [10]**Semiconductor heterogeneous photocatalysis has enormous potential to treat organic contaminants in water and air.** Heterogeneous photocatalytic oxidation has decompose and mineralize recalcitrant organic compounds. The acceleration of photoreaction in the presence of a semiconductor catalyst. TiO_2 , ZnO, Fe_2O_3, CdS, ZnS act as photocatalysts. [11]

TiO_2 ability to break down organic pollutants and even achieve complete mineralization. Photocatalytic and hydrophilic properties of TiO_2 makes it close to an ideal catalyst due to its high reactivity, reduced toxicity, chemical stability and lower costs. Photo electrolysis focused in environmental applications wastewater treatment.[11]

TiO_2 removal of organics as a tertiary treatment employing solar light and 2.3 g/L TiO_2. TOC was reduced from 130 mg/L to 7 mg/L and total elimination of coliforms in less than 60 minutes. TiO_2 photocatalysis with visible irradiation effective for the detoxification of the pharmaceutical salbutamol in water. The most favorable values for drug abatement were 649 mg/L of TiO_2 at a pH of 7.4 irradiated with a 1500 W xenon lamp for 30 minutes which resulted in 93% degradation. [12]

Fenton‘s (Fe^{2+}/H_2O_2) and Fenton‘s-like (Fe^{3+}/H_2O_2) reactions compared for both dark and photo-assisted reactions. After 40 minutes advanced oxidation by Fe^{2+}/H_2O_2 at pH-3, penicillin completely removed. Higher COD and TOC removals obtained with dark Fenton‘s (Fe^{2+}/H_2O_2) at pH-3 compared with dark Fenton‘s-like (Fe^{3+}/H_2O_2). Photo-assisted reactions using UV-C provided only slightly higher removal efficiencies. TOC removal was higher with photo-Fenton‘s reaction and COD removal was slightly higher with photo-Fenton‘s-like reactions. [12]

Penicillin removal by H_2O_2 (30 mM)/UV/ pH 7 22%, Photo-Fenton‘s 56% Photo-Fenton‘s-like 66% Dark Fenton‘s 61%, Dark Fenton‘s-like 46%. Tetracycline and Lincomycin, removal by TiO_2/UV using 98% removal within 2 h both. removal by solar radiation and TiO_2 100% removal 50 mg/L in 200min. 27.4% removal of 5µm initial concentration of ibuprofen, 26.3% of diphenhydramine, 95.8% and 87.8% degradation for phenazone and phenytoin. 110 W low pressure lamp producing monochromatic UV light at 254 nm in 500 mL reactor. [12]

MATERIAL

Hydrogen Peroxide (H_2O_2)

This is the strong oxidant and its application in the treatment of various inorganic and organic pollutants is well established. H_2O_2 consist of two hydrogen molecules and two oxygen molecules.

Fenton's Reagents (Fe salt/ $FeSO_4$ Solution).

Metal salts (e.g. iron salts) which are strong oxidants that is the Fenton's process. Fe^{+3} and Fe^{+2} are used to oxidation of H_2O_2 which decompose or cause of degradation of waste water. The amount of this Fenton reagent is based on the amount used of H_2O_2.

Acid or Alkali

H_2SO_4 acid or NaOH alkali to be used for Ph maintain of waste water. The optimum Value of pH necessary for the Fenton process.

UV Light

UV light is the oxidizing agent used for the process. 400-450 nm after that rate of degradation reduced. Value of UV intensity should be $\lambda < 450$ nm.

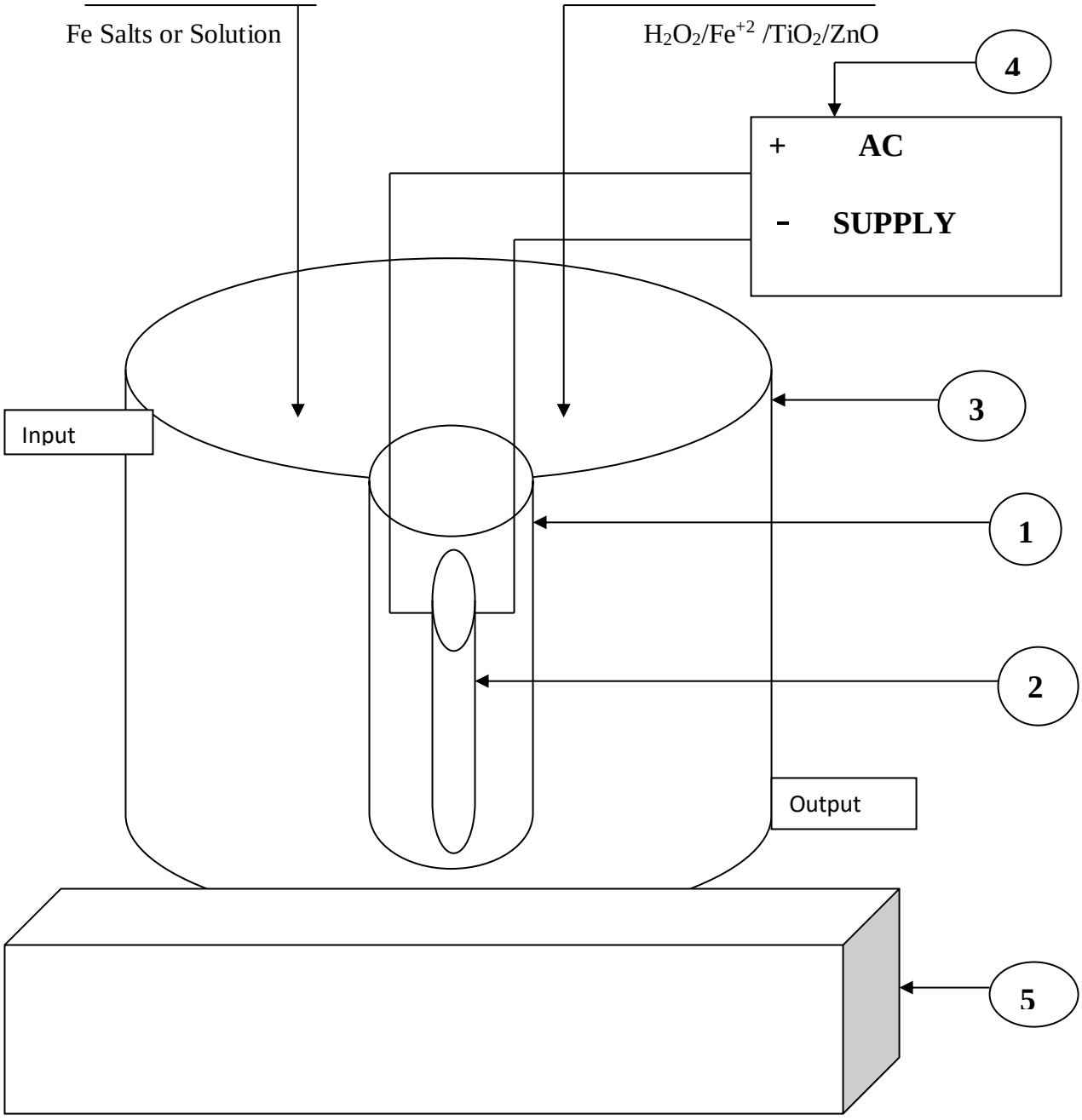
Titanium Oxide (TiO_2)

This is the strong oxidant and its application in the treatment of various inorganic and organic pollutants is well established.

Experimental Setup for Photocatalytic Oxidation Process

Photocatalytic reactor shows by schematic diagram consist of following parts. The reactor can be used for various processes like TiO_2/UV, Fe^{+2}/UV and H_2O_2/UV.

1. UV Lamp Cylinder
2. UV Lamp
3. Cylindrical cell or Reaction Vessel
4. Power supply


Fe Salts or Solution
H_2O_2/Fe^{+2} /TiO_2/ZnO
4
+ AC
- SUPPLY
3
Input
1
2
Output
5

Effect of Contact Time on Paracetamol Reduction Using UV/TiO_2

Contact Time Min	% Removal of Paracetamol For 5 ppm Solution	% Removal of Paracetamol for 10 ppm Solution
30	40	29
60	57	50
90	68	64.5
120	73.3	69
150	80	72.5

Effect of Contact Time on Paracetamol Reduction Using UV/TiO_2

Table No. 6.1 shows the effect of contact time on % reduction of paracetamol using UV/TiO_2 process. The experimental analysis carried out for 5 ppm and 10 ppm solutions. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 40, 57, 68, 73 and 80 resp. using UV/TiO_2 process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 29, 50, 64,69 and 72.5 resp. using UV/TiO_2 process. As per analysis its clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time

Effect of Contact Time on Paracetamol Reduction Using UV/ZnO Table No. 6.2 shows the effect of contact time on % reduction of paracetamol using UV/ZnO process. The experimental analysis carried out for 5 ppm and 10 ppm solutions. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 30, 50, 62.5, 71.5 and 77 resp. using UV/ZnO process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs are 25, 42, 57.5 and 69 resp. using UV/ZnO process. As per analysis it's clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

Contact Time Min	% Removal of Paracetamol For 5 ppm Solution	% Removal of Paracetamol for 10 ppm Solution
30	30	25
60	50	42

90	62.5	57
120	71.5	64.5
150	77	69

Effect of Contact Time on Paracetamol Reduction Using UV/ZnO

Effect Parameters on Rate of Degradation [1]

Effect Parameters on Degradation Paracetamol & Procaine TiO_2

1. Effect of Catalyst Weight

The effect of TiO_2 catalyst amount on the degradation of paracetamol and procaine solutions at natural pH. Typical weight of TiO_2 5-6 gm/l or 0.02 gm of paracetamol and 0.1 gm for the procaine. Stability the rate of photocatalytic degradation when amount of catalyst reached to 0.05 gm or more for paracetamol and 0.15 gm or more for procaine.

2. Effect of Initial Concentration Paracetamol and Procaine Solutions

The effect of initial concentration for paracetamol and procaine solutions under experimental condition which are 0.02 gm of TiO_2 for paracetamol and 0.1 gm for procaine, 298K and the pH equal 8 & 7 of paracetamol and procaine respectively. Concentration is studied in the range (5- 40 ppm) for both paracetamol and procaine solutions.

3. Effect of pH

pH is an important factor in evaluation of photocatalytic degradation rate because the pH change affects the adsorption quantity of organic pollutants. Photocatalytic degradation rate optimal at pH is 8.0 for paracetamol and 4.0 for procaine in presence of certain conditions. pH effect range (4, 6, 8, 10 and 12) for both paracetamol and procaine.

4. Effect of Temperature

The study of temperature effect on photocatalytic degradation rate optimal temperature 55 ^{O}C for paracetamol and procaine with initial concentration 10µg/ml for both paracetamol and procaine TiO_2 amount 5-6 gm/L or 0.02 gm for paracetamol and 0.1 gm for procaine & pH solution 8 & 4 for paracetamol & procaine.

4. Effect of Contact Time

As per analysis it's clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

Effect Parameters On Degradation of Tetracycline by ZnO

1. Effect of Mass Dosage

The effect of catalyst loading on the last De-colonization capacity a chains of data carry out via changing the substance about 0.5-3.0 g/l in the solution by 50 mg/l concentration drug, at time one hour and temp. 25 ^{O}C. Photocatalytic degradation rate of photo degradation raise via raising the quantity of ZnO equal to 0.2 g/l. Photo catalytic degradation was taken a plateau region of 0.05- 0.3 g/L then the rate of degradation lowering by raise in weight dose.

2. The Effect of Primary Concentration of Drug

The removal of drug has been conducted via utilizing several primary concentration drug about (10 - 80 mg/L). At series (0.2 gm / 100 cm^3) the suspension solution was irradiated by 7.33 m W/cm^2 light intensity, flow rate of air bubble 10 cm3/ min, at 25oC and 0.2 gm/100 cm3 of ZnO Nano particle as actuals. The best drug concentration was 50 mg/L the utmost drug removal because the drug was cover the major area of the ZnO Nano particle, thus absorbed best exciting photons to generate greater conc. of the activated catalyst.

3. Effect of pH

pH is an important factor in evaluation of photocatalytic degradation rate because the pH change affects the adsorption quantity of organic pollutants. Photocatalytic degradation rate optimal at pH is 8.0 for paracetamol and 4.0 for procaine in presence of certain conditions. pH effect range (4, 6, 8, 10 and 12) for Tetracycline.

CONCLUSIONS

Both UV light and catalyst are important factors in paracetamol degradation. The initial concentration increase for both drugs leads to reduce degradation rate with foundation of the mentioned weights of ZnO/TiO_2. Parameters that effects on photocatalytic degradation of Pharmaceutical wastewater are initial effluent concentration, catalyst dosage, irradiation time, intensity of UV light and effect of pH. When the amount of Photocatalyst load increased in the wastewater photocatalytic degradation efficiency significantly increase. The degradation of pharmaceuticals is highly dependent on the operational parameters of the system. Operating parameters such as initial pH, initial concentration of the pollutant, catalyst loading and irradiation time can influence the removal rate of pollutants. The photocatalytic degradation able to removal of pharmaceutical compounds such as ketoprofen, ibuprofen, tetracycline, amoxicillin and naproxen. The three catalysts (Titanium dioxide, zinc oxide and TiO_2/ H_2O_2) used are effective catalysts in photocatalytic degradation of real pharmaceutical wastewater.

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