Removal of Paracetamol Drugs from Pharmaceutical Wastewater Using TiO₂/Solar and ZnO/Solar

Mrs. Shital Dond¹ and Dr. S.A. Misal²

1P.G. Students of Department of Chemical Engineering and 2Head of Department, Chemical Engineering Pravara Rural Engineering College, Loni, Dist.: Ahmednagar- 413736.

ABSTRACT

The effect of concentration on % reduction of paracetamol using ZnO/Solar and TiO₂/Solar process. For concentrations 5, 10, 15, 20, 25 and 30 ppm solutions % reduction of paracetamol using ZnO/Solar process are 78, 70, 65, 61, 54, 51 and using TiO₂/solar process are 80, 73, 68, 64, 57, 54 resp. for 120-150 contact time. As per observation as the concentration of paracetamol increase, rate of reduction decrease. As per observations both the processes are economical and having same efficiency for paracetamol reduction. For TiO₂/Solar Process gives more reduction of drugs than the ZnO/Solar process. There is little bit difference in reduction of paracetamol between two processes. Both the processes are effective for paracetamol reduction. As the concentration of pollutant increase the rate of reduction decreases. As per observation for paracetamol drugs from synthetic wastewater is 72.5 and 69.44% at 150 min time with 10 ppm concentration of dugs for TiO₂/Solar process resp.

Keywords –TiO2/Solar Process, ZnO/Solar Process, Reduction of Paracetamol, Concentration, pH, Contact Time.

Introduction

Due to rapid growth of population and industrialization the requirement of water increases 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. Goal of biological wastewater treatment is stepwise oxidation of organic pollutants.

Materials

- 1. Hydrogen Peroxide (H₂O₂)
- 2. Titanium Oxide (TiO₂)
- 3. Acid or Alkali
- 4. Solar Light
- 5. Zinc Oxide (ZnO)

Chemicals and Reagents

- 1. ZnO
- $2.\ TiO_2$
- 3. 0.1 N NaOH or H₂SO₄
- 4. Paracetamol
- 5. Distilled Water

Experimental Process for various Contact Time

- 1. Take 0.5 L of 10 ppm pharmaceutical wastewater in beaker and stirred for mixing.
- 2. Measure the absorbance of known 10 ppm solution or wastewater.
- 3. Add 1 gm/ L of ZnO/TiO₂ catalyst in beaker.
- 4. Stirred the solution for 10 min.
- 5. Keep the beakers of various concentrations solutions in sunlight.
- 6. For various time of interval 30, 60, 90, 120 and 150-min withdrawal of sample.
- 7. Check absorbance unit for each sample wastewater help of colorimeter.

8. Calculate % reduction of various for paracetamol by comparing initial and final absorbance unit taken using colorimeter.

11. Follow this procedure for various pH for 10 ppm solution and calculate the % reduction for the paracetamol.

Experimental Process for various Concentration

1. Take 0.5 L of 5 ppm pharmaceutical wastewater in reactor and stirred for mixing.

- 2. Measure the absorbance of known 5 ppm solution or wastewater.
- 3. Add 1 gm/ L of ZnO/TiO₂ catalyst in beaker.
- 4. Stirred the solution for 10 min.
- 5. Follow the same procedure for 10, 15, 20 and 25 ppm synthetic wastewater solution.
- 6. Measure the absorbance for above various ppm solution.
- 7. Keep the beakers of various concentrations solutions in sunlight.
- 8. For contact time of interval 90-min withdrawal of sample.

9. Check absorbance unit for each sample wastewater help of colorimeter.

10. Calculate % reduction of various for paracetamol by comparing initial and final absorbance unit taken using colorimeter.

Experimental Process for various pH

- 1. Take 0.5 L of 10 ppm pharmaceutical wastewater in beaker
- 2. Stirred for mixing.
- 3. Stirred the solution for 10 min.
- 4. Measure pH for solution.

5. If pH less than 7 we can increase by adding 0.1 N NaOH solution and pH more than 7 we can reduce by adding 0.1 N H_2SO_4 solution.

6. Make the various pH solutions 2, 4, 6, 8 and 10.

7. Add 1 gm/ L of ZnO/TiO₂ catalyst in beaker.

8. Keep the beakers of various concentrations solutions in sunlight.

9. For 90 min contact time withdrawal of sample.

10. Check absorbance unit for each sample wastewater help of colorimeter.

% Reduction of Paracetamol Using TiO₂/Solar



Fig. Graphical Representation % Reduction of Paracetamol for TiO₂/Solar

For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins % reduction for paracetamol drugs is 40, 57, 68, 73 and 80 resp. using TiO₂/Solar process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins % reduction for paracetamol drugs is 29, 50, 64,69 and 72.5 resp. using TiO₂/Solar process. % Reduction increases with increase in contact time. The max. reduction shown at 120-150 mins.

Graphical Representation of % Paracetamol Reduction

For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins % reduction for paracetamol drugs is 33.3, 50, 62.5, 71.5 and 77.7 resp. using ZnO/Solar process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins % reduction for paracetamol drugs is 21, 42, 58, 64.5 and 70 resp. using ZnO/Solar process. 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.





Graphical Representation of % Reduction of Paracetamol 5 ppm



Fig. Graphical Representation of % Reduction of Paracetamol 5 ppm

For TiO₂/Solar _{Process} gives more reduction of drugs than the ZnO/Solar process. There is little bit difference in reduction of paracetamol between two processes. Both the processes are effective for paracetamol reduction.

% Reduction of Paracetamol 10 ppm

Fig. shows the comparison of two AOP for reduction of paracetamol drugs from synthetic wastewater for 10 ppm at various contact time. For $TiO_2/Solar$ Process gives more reduction of drugs than the ZnO/Solar process. There is little bit difference in reduction of paracetamol between two processes.



Fig. Graphical Representation of % Reduction of Paracetamol 10 ppm

% Reduction of Paracetamol for ZnO/Solar and TiO₂/Solar at various concentrations



Fig. Graphical Representation % Reduction of Paracetamol for TiO_/Solar

% reduction of paracetamol using ZnO/Solar and TiO₂/Solar process. As per graphical representation, as the concentration of paracetamol increase, rate of reduction decrease. As per observations both the processes are economical and having same efficiency for paracetamol reduction.

Effect Parameters on Rate of Degradation

1. Effect of Catalyst Weight

The effect of TiO_2 and ZnO catalyst amount on the degradation of paracetamol and procaine solutions at natural ph. Typical weight of TiO_2 and ZnO 1-5 gm/l for 5-50 ppm paracetamol solution. Stability the rate of photocatalytic degradation when amount of catalyst reached to 05 gm/l 25-30 ppm solution.

2. Effect of Initial Concentration Paracetamol Solutions

The effect of initial concentration for paracetamol solutions is important in terms of % reduction. Concentration is studied in the range (5-30 ppm) for both processes TiO₂/solar and ZnO/solar paracetamol 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 paracetamols.

4. Effect of Temperature

The study of temperature effect on photocatalytic degradation rate optimal temperature 55-60 $^{\circ}$ C for paracetamol with initial concentration 10 mg/l for both processes TiO₂/solar and ZnO/solar & pH solution 6-8 for paracetamol.

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.

CONCLUSIONS

Effect on photocatalytic degradation of pharmaceutical wastewater is initial effluent concentration, catalyst dosage, irradiation time, intensity of solar light and effect of pH. Photocatalyst load increased in the wastewater photocatalytic degradation efficiency significantly increase. Concentration is the important parameter that impact on the rate of reduction of paracetamol from the wastewater. Catalyst (ZnO and TiO₂) use for treatment is fixed which is 1-6 gm/l using solar radiation as light source. As pH of paracetamol in wastewater increase the rate of reduction also increase. pH of paracetamol in wastewater be the important parameter that impact on the rate of paracetamol from the wastewater. For alkaline pH solution the rate of paracetamol reduction using ZnO/solar process will be maximum. 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. Titanium dioxide, zinc oxide are effective catalysts in photocatalytic degradation of real pharmaceutical wastewater.

REFERENCES

1. Aseel M. Aljeboree and Firas H. Abdulrazzak, Photocatalytic of Pharmaceutical Tetracycline (TCS) By Zinc Oxide (ZnO) College of Science for Women-Chemistry Department/ University of Babylon-Iraq. 2University of Diyala, Baqubah, Iraq, Journal of Critical Reviews ISSN-2394-5125 Vol 7, Issue 7, 2020.

2. Alaa Jawad Abdul-Zahra, Hanaa Addai Al-Sultani, Muthanna Saleh Mashkour and Amer Muosa Juda, Photocatalytic degradation of paracetamol and procaine, Branch of chemistry, Faculty of pharmacy and Chemistry Department, Faculty of Science, University of Kufa, Najaf, Iraq, International Journal of Chemtech Research CODEN (USA): IJCRGG, ISSN: 0974-4290, ISSN(Online), 2455-9555 Vol.9, No.11 pp 412-425, 2016.

3. Ankit M. Bhuva and Darshana T. Bhatti, Photocatalytic Degradation of Pharmaceutical Compounds Using Titanium Dioxide Nano Particles, Department of Chemical Engineering, V.V.P. Engineering College, Rajkot, Gujarat, India, International Journal of Advance Engineering and Research Development Volume 2, Issue 2, February -2015.

4. G. Suvarna Lakshmi and M. V. V. Chandana Lakshmi, Removal of Organic Pollutants from the Pharmaceutical Effluent by Tio₂ Based Photocatalysis, M. Tech, Department of Chemical Engineering, AUCOE (A), Andhra University, Visakhapatnam, India1 Professor, Department of Chemical Engineering, AUCOE (A), Andhra University, Visakhapatnam, India, International Journal of Innovative Research in Science, Engineering and Technology, Vol. 5, Issue 8, August 2016.

5.Khasawneh Omar and Puganeshwary Palaniandy, Photocatalytic Degradation of Pharmaceuticals Using TiO₂ Based Nanocomposite Catalyst- Review, School of Civil Engineering Campus, University Sains Malaysia, Malaysia, Civil and Environmental Engineering Reports, E-ISSN 2450-8594 CEER 2019; 29 (3): 001-033.

6. Kumudini V. Marathe, Rathod, Chandrakant Gadipelly and Ganapati D. Yadav, Department of Chemical and Biomolecular Engineering, University of Cantabria, Cantabria 39005, Spain Industrial & Engineering Chemistry Research and Pharmaceutical Industry Wastewater: Review of the Technologies for Water Treatment and Reuse Department of Chemical Engineering Institute of Chemical Technology, Mumbai 400019, India.

7. Khan Z Wasi, Photodegradation of Real Pharmaceutical Wastewater with Titanium Dioxide, Zinc Oxide, and Hydrogen Peroxide During UV Treatment Military Technological College, Muscat, Oman, IOSR Journal of Engineering (IOSRJEN), ISSN (e): 2250-3021, ISSN (p): 2278-8719 Vol. 06, Issue 07 (July. 2016), PP 36-46.

8. Klaus Kuhn, Joachim Fauler and Sara Teixeira, Photocatalytic degradation of pharmaceuticals present in conventional treated wastewater by nanoparticle suspensions, Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 and Institute of Clinical Pharmacology, Faculty of

Medicine Carl Gustav Carus, TU Dresden, 01307 Dresden, Germany, Journal of Environmental Chemical Engineering.

9. Melati Ireng Sari and Tuty Emilia Agustina, Color and COD Degradation in Photocatalytic Process of Procion Red by Using TiO₂ Catalyst under Solar Irradiation, Graduate Program Student Chemical Engineering Magister Program and Engineering Faculty, Universitas Sriwijaya, Indonesia Chemical Engineering Department, Engineering Faculty, Universitas Sriwijaya, Indonesia.

Malato Sixto, Waste water treatment by advanced oxidation processes (solar photocatalysis in degradation of industrial contaminants), Plataforma Solar de Almería-CIEMAT. Carretera Sense km4, Tabernas (Almería).
O4200- Spain, Innovative Processes and Practices for Wastewater Treatment and Re-use 8-11 October 2007, Ankara University, Turkey.

11.Muhammad Umar and Hamidi Abdul Aziz, Photocatalytic Degradation of Organic Pollutants in Wate, Organic Pollutants - Monitoring, Risk and Treatment, Photocatalytic Degradation of Organic Pollutants in Water, http://dx.doi.org/10.5772/53699.

12. M. Gandhirajan, G. Amarnath, P. Kavitha and Rakhee Bhagavath, Characterization and Treatment of Pharmaceutical R&D Wastewater, Tech-Sharp Enviro Systems (P) Ltd., C-39, Second Avenue, Anna Nagar, Chennai 600 040, T.N., India, Journal of Industrial Pollution Control 24 (1) (2008) pp 1-8.

13. Marie Deegan, Pharmaceuticals in industrial wastewater and their removal using photo-Fenton 's oxidation, Ph.D. Research Thesis, School of Biotechnology Dublin City University Dublin 9 Ireland July 2011.

14. Navaneetha, B. Venkateswara Reddy, P. Sandeep, P. Ujwala & K. Venkata Ramana Reddy, Water Treatment Process in Pharma Industry - A Review, Department of Pharmaceutics, St. Paul's college of Pharmacy, Turkayamjal (V), Hayathnagar (M), Rangareddy Dist-501510, Andhra Pradesh and Sree Datta collage of pharmacy, Sheriguda (v), International Journal of Pharmacy and Biological Sciences (e-ISSN: 2230-7605) IJPBS, Volume 4I Issue 2, APR-JUN, 2014, 07-18.

15. Paul Sebastian, Baskaran S, Sadish Oumabady, Davamani V and Kalaiselvi P, Integrated Advanced Oxidation Process for the Treatment of Pharmaceutical Effluent, Madras Agric. J., 2021.

16. S. Mahamood and B. Manu, Enhanced degradation of paracetamol by UV-C supported photo-Fenton process over Fenton oxidation, Water Science & Technology, 64.12, 2434, 2011.

17. Saranya R and Nithyanandhi M, Removal of Paracetamol from Pharmaceutical Wastewater by Integrated MBR, Journal of Chemical and Pharmaceutical Sciences ISSN: 0974-2115, February, 2017.

18. Santos Naiara de Oliveira dos, Teixeira Luiz A.C., Jianan Li, Chaoran Li and Luiza C. Campos, Removal of diethyltoluamide, paracetamol, caffeine and triclosan from natural water by photo-Fenton process using powdered zero-valent iron, Journal of Water Process Engineering 48, 102907, 2022.

19. Sanromán, M. A., Meijide, J., Lama, G., Pazos, M., & Dunlop, P. S. M. (2022). Ultraviolet-based heterogeneous advanced oxidation processes as technologies to remove pharmaceuticals from wastewater: An overview. Journal of Environmental Chemical Engineering, 10, 2022. (107630. https://doi.org/10.1016/j.jece.2022.107630).

20. Selvasembian Rangabhashiyam, Deborah Tebogo Ruziwa, Abimbola E. Oluwalana, Mathew Mupa, Lucas Meili and Matthew M. Nindi, Pharmaceuticals in wastewater and their photocatalytic degradation using nanoenabled photocatalysts, Journal of Water Process Engineering 54, 103880, 2023.